Supporting Information

Nan Zhang,[a] Chunli Liu,[b,c,d] Jinghong Ma,[a] Ruifeng Li,[a] Haijun Jiao*[b,e]*

(a) College of Chemistry and Chemical Engineering, Taiyuan University of Technology, Taiyuan, 030024, China. b) State Key Laboratory of Coal Conversion, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan, 030001, China; c) National Energy Center for Coal to Liquids, Synfuels China Co., Ltd, Huairou District, Beijing, 101400, China; d) University of Chinese Academy of Sciences, No. 19A Yuquan Road, Beijing, 100049, China; e) Leibniz-Institut für Katalyse e.V. an der Universität Rostock, Albert-Einstein Strasse 29a, 18059 Rostock, Germany E-mail: rfli@tyut.edu.cn; haijun.jiao@catalysis.de

Contents

Table S1: Computed electronic energies (eV) of all 48 acidic sites for one Al substitution

T1	T^{Al}_{1}-O^{H}-T^{Si}_{2}	T^{Al}_{1}-O^{H}-T^{Si}_{10}	T^{Al}_{1}-O^{H}-T^{Si}_{5}	T^{Al}_{1}-O^{H}-T^{Si}_{4}
E	-2296.63	-2296.54	-2296.53	-2296.34
T2	T^{Al}_{2}-O^{H}-T^{Si}_{6}	T^{Al}_{2}-O^{H}-T^{Si}_{1}	T^{Al}_{2}-O^{H}-T^{Si}_{8}	T^{Al}_{2}-O^{H}-T^{Si}_{3}
E	-2296.68	-2296.61	-2296.54	-2296.49
T3	T^{Al}_{3}-O^{H}-T^{Si}_{4}	T^{Al}_{3}-O^{H}-T^{Si}_{6}	T^{Al}_{3}-O^{H}-T^{Si}_{12}	T^{Al}_{3}-O^{H}-T^{Si}_{2}
E	-2296.67	-2296.65	-2296.63	-2296.61
T4	T^{Al}_{4}-O^{H}-T^{Si}_{7}	T^{Al}_{4}-O^{H}-T^{Si}_{5}	T^{Al}_{4}-O^{H}-T^{Si}_{1}	T^{Al}_{4}-O^{H}-T^{Si}_{3}
E	-2296.68	-2296.64	-2296.57	-2296.51
T5	T^{Al}_{5}-O^{H}-T^{Si}_{11}	T^{Al}_{5}-O^{H}-T^{Si}_{6}	T^{Al}_{5}-O^{H}-T^{Si}_{1}	T^{Al}_{5}-O^{H}-T^{Si}_{4}
E	-2296.58	-2296.57	-2296.57	-2296.45
T6	T^{Al}_{6}-O^{H}-T^{Si}_{2}	T^{Al}_{6}-O^{H}-T^{Si}_{3}	T^{Al}_{6}-O^{H}-T^{Si}_{5}	T^{Al}_{6}-O^{H}-T^{Si}_{9}
E	-2296.63	-2296.60	-2296.58	-2296.51
T7	T^{Al}_{7}-O^{H}-T^{Si}_{11}	T^{Al}_{7}-O^{H}-T^{Si}_{8}	T^{Al}_{7}-O^{H}-T^{Si}_{4}	T^{Al}_{7}-O^{H}-T^{Si}_{7}
E	-2296.64	-2296.62	-2296.62	-2296.58
T8	T^{Al}_{8}-O^{H}-T^{Si}_{12}	T^{Al}_{8}-O^{H}-T^{Si}_{7}	T^{Al}_{8}-O^{H}-T^{Si}_{9}	T^{Al}_{8}-O^{H}-T^{Si}_{2}
E	-2296.73	-2296.59	-2296.50	-2296.46
T9	T^{Al}_{9}-O^{H}-T^{Si}_{10}	T^{Al}_{9}-O^{H}-T^{Si}_{9}	T^{Al}_{9}-O^{H}-T^{Si}_{6}	T^{Al}_{9}-O^{H}-T^{Si}_{8}
E	-2296.69	-2296.54	-2296.53	-2296.47
T10	T^{Al}_{10}-O^{H}-T^{Si}_{1}	T^{Al}_{10}-O^{H}-T^{Si}_{10}	T^{Al}_{10}-O^{H}-T^{Si}_{11}	T^{Al}_{10}-O^{H}-T^{Si}_{9}
E	-2296.62	-2296.53	-2296.39	-2296.35
T11	T^{Al}_{11}-O^{H}-T^{Si}_{5}	T^{Al}_{11}-O^{H}-T^{Si}_{7}	T^{Al}_{11}-O^{H}-T^{Si}_{12}	T^{Al}_{11}-O^{H}-T^{Si}_{10}
E	-2296.62	-2296.59	-2296.58	-2296.46
T12	T^{Al}_{12}-O^{H}-T^{Si}_{8}	T^{Al}_{12}-O^{H}-T^{Si}_{12}	T^{Al}_{12}-O^{H}-T^{Si}_{3}	T^{Al}_{12}-O^{H}-T^{Si}_{11}
E	-2296.70	-2296.63	-2296.62	-2296.58

Table S2: Bader charge (BC) for H atom in the bare HZSM5 structures

T1	T_1^{Al}-O^H-T_2^{Si}	T_1^{Al}-O^H-T_{10}^{Si}	T_1^{Al}-O^H-T_5^{Si}	T_1^{Al}-O^H-T_4^{Si}
BC/bare HZSM-5	0.67	0.68	0.67	0.65
T2	T_2^{Al}-O^H-T_6^{Si}	T_2^{Al}-O^H-T_1^{Si}	T_2^{Al}-O^H-T_8^{Si}	T_2^{Al}-O^H-T_3^{Si}
BC/bare HZSM-5	0.64	0.70	0.68	0.72
T3	T_3^{Al}-O^H-T_4^{Si}	T_3^{Al}-O^H-T_6^{Si}	T_3^{Al}-O^H-T_{12}^{Si}	T_3^{Al}-O^H-T_2^{Si}
BC/bare HZSM-5	0.72	0.71	0.71	0.69
T4	T_4^{Al}-O^H-T_7^{Si}	T_4^{Al}-O^H-T_5^{Si}	T_4^{Al}-O^H-T_1^{Si}	T_4^{Al}-O^H-T_3^{Si}
BC/bare HZSM-5	0.68	0.67	0.67	0.68
T5	T_5^{Al}-O^H-T_{11}^{Si}	T_5^{Al}-O^H-T_6^{Si}	T_5^{Al}-O^H-T_1^{Si}	T_5^{Al}-O^H-T_4^{Si}
BC/bare HZSM-5	0.66	0.70	0.68	0.66
T6	T_6^{Al}-O^H-T_2^{Si}	T_6^{Al}-O^H-T_3^{Si}	T_6^{Al}-O^H-T_5^{Si}	T_6^{Al}-O^H-T_9^{Si}
BC/bare HZSM-5	0.68	0.68	0.71	0.67
T7	T_7^{Al}-O^H-T_{11}^{Si}	T_7^{Al}-O^H-T_8^{Si}	T_7^{Al}-O^H-T_4^{Si}	T_7^{Al}-O^H-T_7^{Si}
BC/bare HZSM-5	0.67	0.67	0.67	0.67
T8	T_8^{Al}-O^H-T_{12}^{Si}	T_8^{Al}-O^H-T_7^{Si}	T_8^{Al}-O^H-T_9^{Si}	T_8^{Al}-O^H-T_2^{Si}
BC/bare HZSM-5	0.69	0.70	0.68	0.67
T9	T_9^{Al}-O^H-T_{10}^{Si}	T_9^{Al}-O^H-T_9^{Si}	T_9^{Al}-O^H-T_6^{Si}	T_9^{Al}-O^H-T_8^{Si}
BC/bare HZSM-5	0.70	0.69	0.67	0.69
T10	T_{10}^{Al}-O^H-T_1^{Si}	T_{10}^{Al}-O^H-T_{10}^{Si}	T_{10}^{Al}-O^H-T_{11}^{Si}	T_{10}^{Al}-O^H-T_9^{Si}
BC/bare HZSM-5	0.70	0.70	0.66	0.68
T11	T_{11}^{Al}-O^H-T_5^{Si}	T_{11}^{Al}-O^H-T_7^{Si}	T_{11}^{Al}-O^H-T_{12}^{Si}	T_{11}^{Al}-O^H-T_{10}^{Si}
BC/bare HZSM-5	0.66	0.64	0.67	0.65
T12	T_{12}^{Al}-O^H-T_8^{Si}	T_{12}^{Al}-O^H-T_{12}^{Si}	T_{12}^{Al}-O^H-T_3^{Si}	T_{12}^{Al}-O^H-T_{11}^{Si}
BC/bare HZSM-5	0.68	0.71	0.68	0.68

Table S3: Computed electronic energies (eV) of pyridine adsorption at all 48 acidic sites for one Al substitution

T1	T^{Al}_{1}-O^{H}-T^{Si}_{2}	T^{Al}_{1}-O^{H}-T^{Si}_{10}	T^{Al}_{1}-O^{H}-T^{Si}_{5}	T^{Al}_{1}-O^{H}-T^{Si}_{4}
E/Pyridine	-2370.13	-2370.00	-2370.04	-2368.43
T2	T^{Al}_{2}-O^{H}-T^{Si}_{6}	T^{Al}_{2}-O^{H}-T^{Si}_{1}	T^{Al}_{2}-O^{H}-T^{Si}_{8}	T^{Al}_{2}-O^{H}-T^{Si}_{3}
E/Pyridine	-2369.82	-2370.03	-2369.42	-2369.96
T3	T^{Al}_{3}-O^{H}-T^{Si}_{4}	T^{Al}_{3}-O^{H}-T^{Si}_{6}	T^{Al}_{3}-O^{H}-T^{Si}_{12}	T^{Al}_{3}-O^{H}-T^{Si}_{2}
E/Pyridine	-2369.88	-2370.02	-2370.07	-2370.05
T4	T^{Al}_{4}-O^{H}-T^{Si}_{7}	T^{Al}_{4}-O^{H}-T^{Si}_{5}	T^{Al}_{4}-O^{H}-T^{Si}_{1}	T^{Al}_{4}-O^{H}-T^{Si}_{3}
E/Pyridine	-2369.68	-2369.87	-2368.47	-2369.90
T5	T^{Al}_{5}-O^{H}-T^{Si}_{11}	T^{Al}_{5}-O^{H}-T^{Si}_{6}	T^{Al}_{5}-O^{H}-T^{Si}_{1}	T^{Al}_{5}-O^{H}-T^{Si}_{4}
E/Pyridine	-2368.37	-2370.03	-2369.68	-2369.67
T6	T^{Al}_{6}-O^{H}-T^{Si}_{2}	T^{Al}_{6}-O^{H}-T^{Si}_{3}	T^{Al}_{6}-O^{H}-T^{Si}_{5}	T^{Al}_{6}-O^{H}-T^{Si}_{9}
E/Pyridine	-2369.56	-2369.17	-2369.86	-2369.84
T7	T^{Al}_{7}-O^{H}-T^{Si}_{11}	T^{Al}_{7}-O^{H}-T^{Si}_{8}	T^{Al}_{7}-O^{H}-T^{Si}_{4}	T^{Al}_{7}-O^{H}-T^{Si}_{7}
E/Pyridine	-2370.13	-2370.07	-2370.11	-2370.09
T8	T^{Al}_{8}-O^{H}-T^{Si}_{12}	T^{Al}_{8}-O^{H}-T^{Si}_{7}	T^{Al}_{8}-O^{H}-T^{Si}_{9}	T^{Al}_{8}-O^{H}-T^{Si}_{2}
E/Pyridine	-2368.49	-2370.00	-2369.97	-2369.48
T9	T^{Al}_{9}-O^{H}-T^{Si}_{10}	T^{Al}_{9}-O^{H}-T^{Si}_{9}	T^{Al}_{9}-O^{H}-T^{Si}_{6}	T^{Al}_{9}-O^{H}-T^{Si}_{8}
E/Pyridine	-2369.62	-2369.80	-2369.83	-2369.98
T10	T^{Al}_{10}-O^{H}-T^{Si}_{1}	T^{Al}_{10}-O^{H}-T^{Si}_{10}	T^{Al}_{10}-O^{H}-T^{Si}_{11}	T^{Al}_{10}-O^{H}-T^{Si}_{9}
E/Pyridine	-2370.06	-2370.02	-2368.65	-2369.81
T11	T^{Al}_{11}-O^{H}-T^{Si}_{5}	T^{Al}_{11}-O^{H}-T^{Si}_{7}	T^{Al}_{11}-O^{H}-T^{Si}_{12}	T^{Al}_{11}-O^{H}-T^{Si}_{10}
E/Pyridine	-2369.35	-2370.02	-2370.09	-2369.85
T12	T^{Al}_{12}-O^{H}-T^{Si}_{8}	T^{Al}_{12}-O^{H}-T^{Si}_{12}	T^{Al}_{12}-O^{H}-T^{Si}_{3}	T^{Al}_{12}-O^{H}-T^{Si}_{11}
E/Pyridine	-2369.69	-2369.95	-2370.06	-2369.99

Table S4: Computed electronic energies (eV) of ammonia adsorption at all 48 acidic sites for one Al substitution

T1	T^{Al}_{1}-O^{H}-T^{Si}_{2}	T^{Al}_{1}-O^{H}-T^{Si}_{10}	T^{Al}_{1}-O^{H}-T^{Si}_{5}	T^{Al}_{1}-O^{H}-T^{Si}_{4}
E/NH_3	-2317.63	-2317.60	-2317.63	-2317.31
T2	T^{Al}_{2}-O^{H}-T^{Si}_{6}	T^{Al}_{2}-O^{H}-T^{Si}_{1}	T^{Al}_{2}-O^{H}-T^{Si}_{8}	T^{Al}_{2}-O^{H}-T^{Si}_{3}
E/NH_3	-2317.66	-2317.63	-2317.45	-2317.63
T3	T^{Al}_{3}-O^{H}-T^{Si}_{4}	T^{Al}_{3}-O^{H}-T^{Si}_{6}	T^{Al}_{3}-O^{H}-T^{Si}_{12}	T^{Al}_{3}-O^{H}-T^{Si}_{2}
E/NH_3	-2317.66	-2317.64	-2317.65	-2317.64
T4	T^{Al}_{4}-O^{H}-T^{Si}_{7}	T^{Al}_{4}-O^{H}-T^{Si}_{5}	T^{Al}_{4}-O^{H}-T^{Si}_{1}	T^{Al}_{4}-O^{H}-T^{Si}_{3}
E/NH_3	-2317.75	-2317.82	-2316.60	-2317.74
T5	T^{Al}_{5}-O^{H}-T^{Si}_{11}	T^{Al}_{5}-O^{H}-T^{Si}_{6}	T^{Al}_{5}-O^{H}-T^{Si}_{1}	T^{Al}_{5}-O^{H}-T^{Si}_{4}
E/NH_3	-2317.61	-2317.67	-2317.67	-2317.61
T6	T^{Al}_{6}-O^{H}-T^{Si}_{2}	T^{Al}_{6}-O^{H}-T^{Si}_{3}	T^{Al}_{6}-O^{H}-T^{Si}_{5}	T^{Al}_{6}-O^{H}-T^{Si}_{9}
E/NH_3	-2317.59	-2317.55	-2317.64	-2317.56
T7	T^{Al}_{7}-O^{H}-T^{Si}_{11}	T^{Al}_{7}-O^{H}-T^{Si}_{8}	T^{Al}_{7}-O^{H}-T^{Si}_{4}	T^{Al}_{7}-O^{H}-T^{Si}_{7}
E/NH_3	-2317.72	-2317.72	-2317.72	-2317.72
T8	T^{Al}_{8}-O^{H}-T^{Si}_{12}	T^{Al}_{8}-O^{H}-T^{Si}_{7}	T^{Al}_{8}-O^{H}-T^{Si}_{9}	T^{Al}_{8}-O^{H}-T^{Si}_{2}
E/NH_3	-2316.81	-2317.63	-2317.58	-2317.50
T9	T^{Al}_{9}-O^{H}-T^{Si}_{10}	T^{Al}_{9}-O^{H}-T^{Si}_{9}	T^{Al}_{9}-O^{H}-T^{Si}_{6}	T^{Al}_{9}-O^{H}-T^{Si}_{8}
E/NH_3	-2316.20	-2317.64	-2317.59	-2317.64
T10	T^{Al}_{10}-O^{H}-T^{Si}_{1}	T^{Al}_{10}-O^{H}-T^{Si}_{10}	T^{Al}_{10}-O^{H}-T^{Si}_{11}	T^{Al}_{10}-O^{H}-T^{Si}_{9}
E/NH_3	-2317.65	-2317.68	-2317.68	-2317.47
T11	T^{Al}_{11}-O^{H}-T^{Si}_{5}	T^{Al}_{11}-O^{H}-T^{Si}_{7}	T^{Al}_{11}-O^{H}-T^{Si}_{12}	T^{Al}_{11}-O^{H}-T^{Si}_{10}
E/NH_3	-2317.65	-2317.64	-2317.71	-2317.54
T12	T^{Al}_{12}-O^{H}-T^{Si}_{8}	T^{Al}_{12}-O^{H}-T^{Si}_{12}	T^{Al}_{12}-O^{H}-T^{Si}_{3}	T^{Al}_{12}-O^{H}-T^{Si}_{11}
E/NH_3	-2316.57	-2317.64	-2317.62	-2317.62

Table S5: Computed electronic energies (eV) of the adsorption of NH_2CH_3, $NH(CH_3)_2$ and $N(CH_3)_3$ at the T1, T3, T5, T7, T11 and T12 centers for one Al substitution

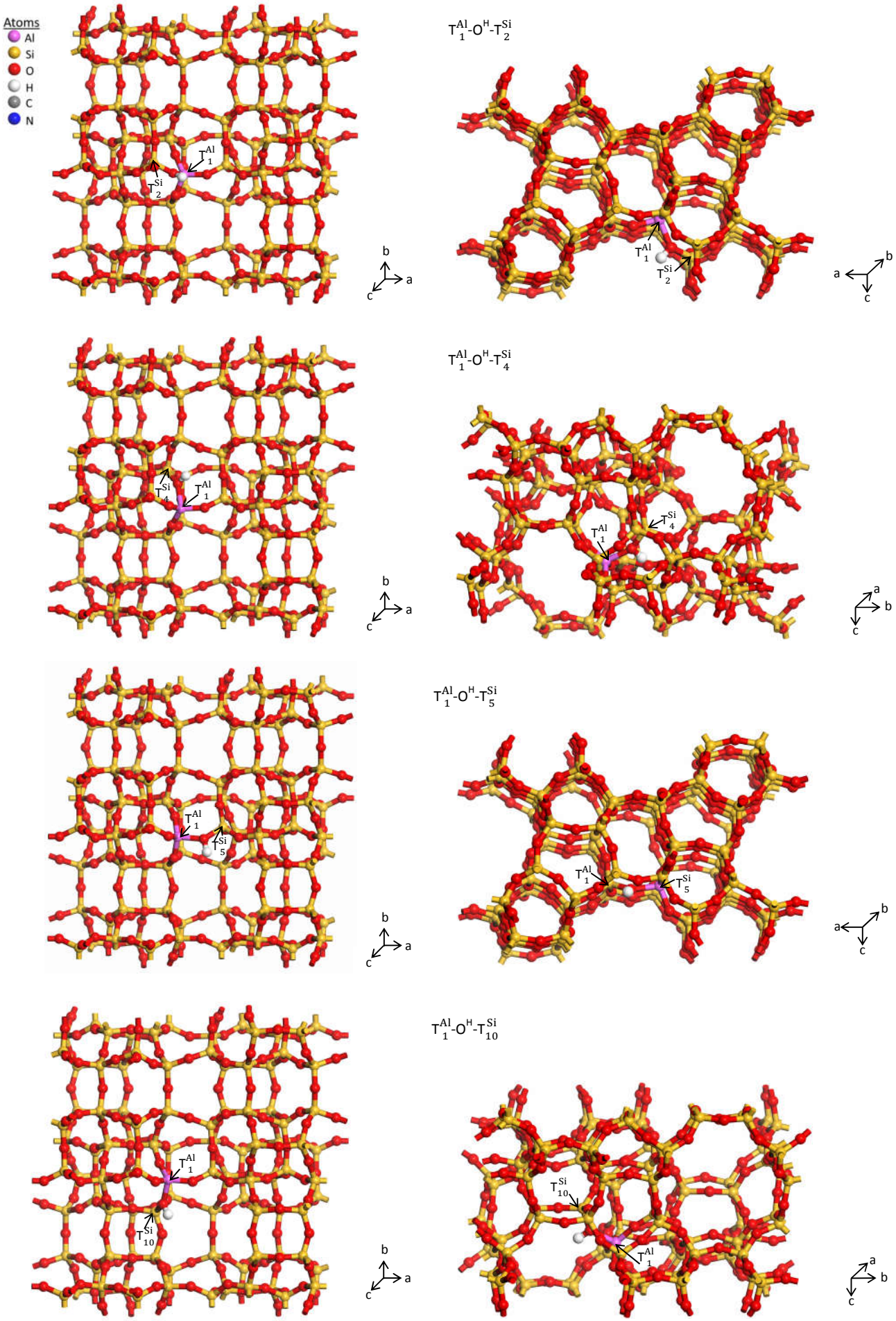
T1	T_1^{Al}-O^H-T_2^{Si}	T_1^{Al}-O^H-T_{10}^{Si}	T_1^{Al}-O^H-T_5^{Si}	T_1^{Al}-O^H-T_4^{Si}
E/NH_2CH_3	-2334.23	-2334.16	-2334.23	-2332.68
E/$NH(CH_3)_2$	-2350.82	-2350.79	-2350.82	-2348.95
E/$N(CH_3)_3$	-2367.28	-2367.21	-2367.10	-2365.55
T3	T_3^{Al}-O^H-T_4^{Si}	T_3^{Al}-O^H-T_6^{Si}	T_3^{Al}-O^H-T_{12}^{Si}	T_3^{Al}-O^H-T_2^{Si}
E/NH_2CH_3	-2334.24	-2334.16	-2334.16	-2334.16
E/$NH(CH_3)_2$	-2350.79	-2350.79	-2350.76	-2350.82
E/$N(CH_3)_3$	-2367.21	-2367.21	-2366.91	-2367.18
T5	T_5^{Al}-O^H-T_{11}^{Si}	T_5^{Al}-O^H-T_6^{Si}	T_5^{Al}-O^H-T_1^{Si}	T_5^{Al}-O^H-T_4^{Si}
E/NH_2CH_3	-2334.15	-2334.21	-2334.16	-2334.18
E/$NH(CH_3)_2$	-2350.76	-2350.82	-2350.78	-2350.80
E/$N(CH_3)_3$	-2366.86	-2367.08	-2367.25	-2367.22
T7	T_7^{Al}-O^H-T_{11}^{Si}	T_7^{Al}-O^H-T_8^{Si}	T_7^{Al}-O^H-T_4^{Si}	T_7^{Al}-O^H-T_7^{Si}
E/NH_2CH_3	-2334.17	-2334.25	-2334.36	-2334.27
E/$NH(CH_3)_2$	-2350.84	-2350.78	-2350.94	-2350.89
E/$N(CH_3)_3$	-2367.22	-2367.22	-2367.29	-2367.11
T11	T_{11}^{Al}-O^H-T_5^{Si}	T_{11}^{Al}-O^H-T_7^{Si}	T_{11}^{Al}-O^H-T_{12}^{Si}	T_{11}^{Al}-O^H-T_{10}^{Si}
E/NH_2CH_3	-2334.04	-2334.06	-2334.23	-2334.05
E/$NH(CH_3)_2$	-2350.78	-2350.83	-2350.85	-2350.20
E/$N(CH_3)_3$	-2366.98	-2367.23	-2367.20	-2365.49
T12	T_{12}^{Al}-O^H-T_8^{Si}	T_{12}^{Al}-O^H-T_{12}^{Si}	T_{12}^{Al}-O^H-T_3^{Si}	T_{12}^{Al}-O^H-T_{11}^{Si}
E/NH_2CH_3	-2332.06	-2334.17	-2334.04	-2334.13
E/$NH(CH_3)_2$	-2348.75	-2350.73	-2350.75	-2350.71
E/$N(CH_3)_3$	-2365.63	-2367.27	-2367.24	-2367.18

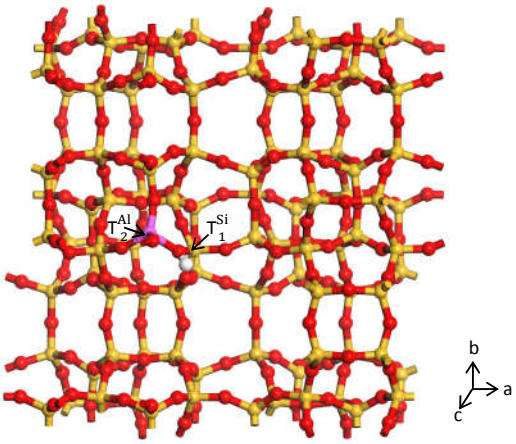
Table S6: All simulated desorption temperature of pyridine and NH_3

T1		A=E11/K	A=E12/K	A=E13/K	A=E14/K
$\Delta H_{ads}/NH_3$	–128.2/–1.33	482	452	425	401
ΔH_{ads}/Pyridine	–204.7/–2.12	760	712	670	631
T2					
$\Delta H_{ads}/NH_3$	–125.6/–1.30	473	442	416	392
ΔH_{ads}/Pyridine	–191.0/–1.98	711	666	626	591
T3					
$\Delta H_{ads}/NH_3$	–128.3/–1.33	484	451	424	400
ΔH_{ads}/Pyridine	–204.5/–2.12	758	712	670	631
T4					
$\Delta H_{ads}/NH_3$	–141.6/–1.47	531	498	468	441
ΔH_{ads}/Pyridine	–175.7/–1.83	654	614	577	545
T5					
$\Delta H_{ads}/NH_3$	–139.0/–1.44	521	488	458	433
ΔH_{ads}/Pyridine	–200.6/–2.08	744	699	656	620
T6					
$\Delta H_{ads}/NH_3$	–128.6/–1.33	483	453	426	402
ΔH_{ads}/Pyridine	–180.0/–1.87	671	629	592	558
T7					
$\Delta H_{ads}/NH_3$	–140.7/–1.46	528	495	464	438
ΔH_{ads}/Pyridine	–203.4/–2.12	756	707	665	629
T8					
$\Delta H_{ads}/NH_3$	–120.5/–1.25	454	426	400	376
ΔH_{ads}/Pyridine	–182.1/–1.89	678	637	598	563
T9					
$\Delta H_{ads}/NH_3$	–122.5/–1.27	462	432	407	382
ΔH_{ads}/Pyridine	–182.0/–1.89	678	634	597	564
T10					
$\Delta H_{ads}/NH_3$	–133.9/–1.39	502	472	442	418
ΔH_{ads}/Pyridine	–178.0/–1.84	663	621	584	552
T11					
$\Delta H_{ads}/NH_3$	–136.1/–1.41	511	479	450	424
ΔH_{ads}/Pyridine	–201.3/–2.09	747	700	659	623
T12					
$\Delta H_{ads}/NH_3$	–126.1/–1.31	475	444	417	394
ΔH_{ads}/Pyridine	–189.2/–1.96	706	662	621	586

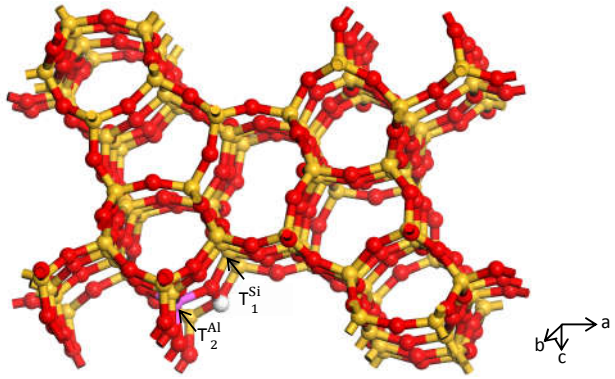
Table S7: Computed electronic energies (eV) of the adsorption of benzene, toluene and p-xylene at the T1, T3, T5, T7, T11 and T12 centers for one Al substitution

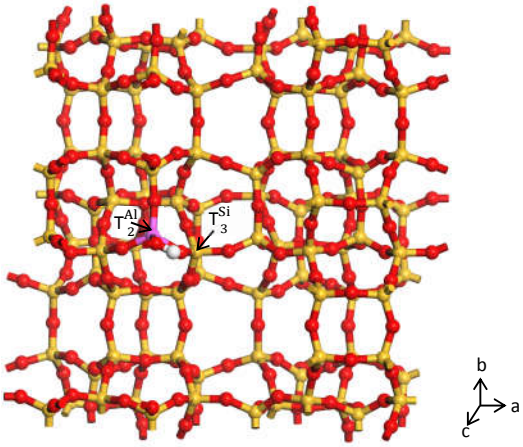
T1	T^{Al}_{1}-O^{H}-T^{Si}_{2}	T^{Al}_{1}-O^{H}-T^{Si}_{10}	T^{Al}_{1}-O^{H}-T^{Si}_{5}	T^{Al}_{1}-O^{H}-T^{Si}_{4}
*E/*benzene	-2373.73	-2373.45	-2373.57	-2372.94
*E/*toluene	-2390.43	-2390.38	-2390.35	-2390.07
*E/*p-xylene	-2407.26	-2406.98	-2407.17	-2406.66
T3	T^{Al}_{3}-O^{H}-T^{Si}_{4}	T^{Al}_{3}-O^{H}-T^{Si}_{6}	T^{Al}_{3}-O^{H}-T^{Si}_{12}	T^{Al}_{3}-O^{H}-T^{Si}_{2}
*E/*benzene	-2373.43	-2373.43	-2373.70	-2373.55
*E/*toluene	-2390.30	-2390.32	-2390.51	-2390.51
*E/*p-xylene	-2406.87	-2406.40	-2407.17	-2407.19
T5	T^{Al}_{5}-O^{H}-T^{Si}_{11}	T^{Al}_{5}-O^{H}-T^{Si}_{6}	T^{Al}_{5}-O^{H}-T^{Si}_{1}	T^{Al}_{5}-O^{H}-T^{Si}_{4}
*E/*benzene	-2373.27	-2373.54	-2373.55	-2373.38
*E/*toluene	-2389.98	-2390.15	-2390.31	-2389.83
*E/*p-xylene	-2406.62	-2406.96	-2407.21	-2406.87
T7	T^{Al}_{7}-O^{H}-T^{Si}_{11}	T^{Al}_{7}-O^{H}-T^{Si}_{8}	T^{Al}_{7}-O^{H}-T^{Si}_{4}	T^{Al}_{7}-O^{H}-T^{Si}_{7}
*E/*benzene	-2373.43	-2373.71	-2373.62	-2373.17
*E/*toluene	-2390.40	-2390.55	-2390.36	-2390.03
*E/*p-xylene	-2407.23	-2407.16	-2407.18	-2406.67
T11	T^{Al}_{11}-O^{H}-T^{Si}_{5}	T^{Al}_{11}-O^{H}-T^{Si}_{7}	T^{Al}_{11}-O^{H}-T^{Si}_{12}	T^{Al}_{11}-O^{H}-T^{Si}_{10}
*E/*benzene	-2373.22	-2373.47	-2373.62	-2373.19
*E/*toluene	-2389.71	-2390.33	-2390.40	-2389.99
*E/*p-xylene	-2406.78	-2407.07	-2406.92	-2406.77
T12	T^{Al}_{12}-O^{H}-T^{Si}_{8}	T^{Al}_{12}-O^{H}-T^{Si}_{12}	T^{Al}_{12}-O^{H}-T^{Si}_{3}	T^{Al}_{12}-O^{H}-T^{Si}_{11}
*E/*benzene	-2372.56	-2373.63	-2373.68	-2373.52
*E/t*oluene	-2390.20	-2390.55	-2390.44	-2390.32
*E/*p-xylene	-2406.78	-2406.89	-2407.16	-2406.94


Atoms
Al
Si
O
H
C
N
T^{Al}_1-O^H-T^{Si}_2
T^{Al}_1-O^H-T^{Si}_4
T^{Al}_1-O^H-T^{Si}_5
T^{Al}_1-O^H-T^{Si}_{10}
b
a
c

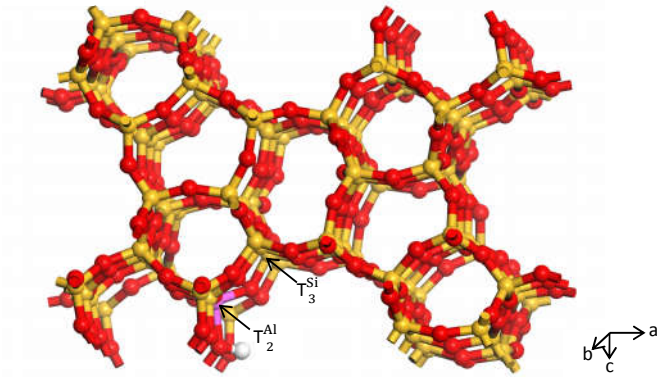


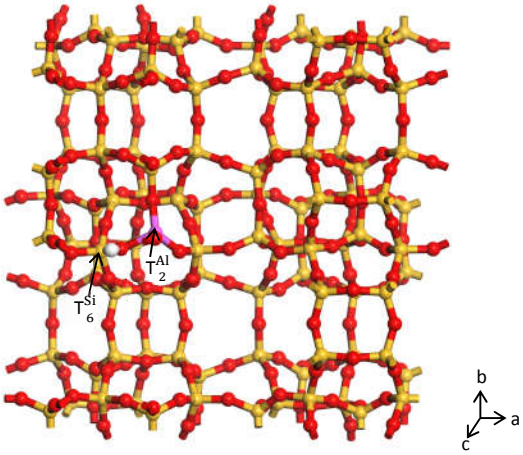
T^{Al}_2-O^H-T^{Si}_1



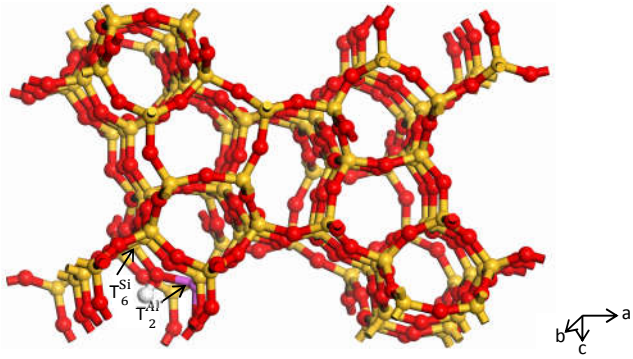


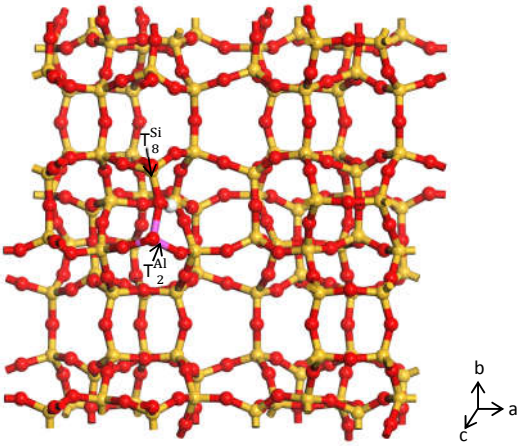
T^{Al}_2-O^H-T^{Si}_3



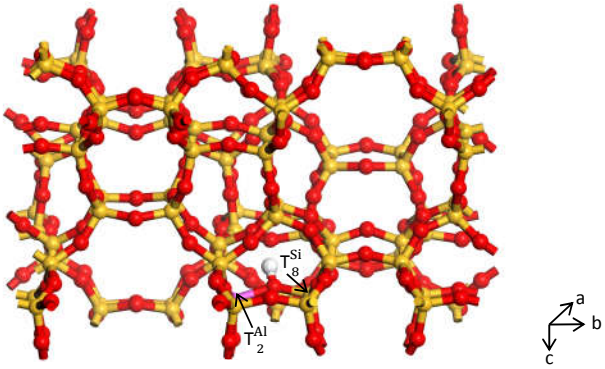


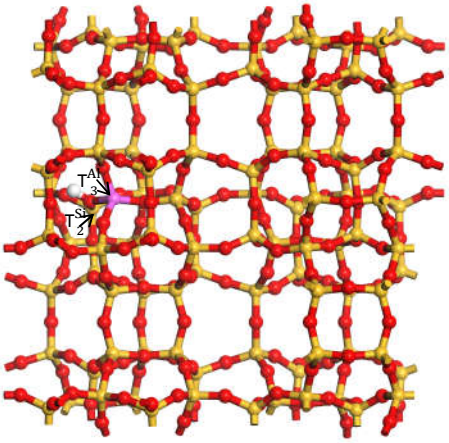
T^{Al}_2-O^H-T^{Si}_6



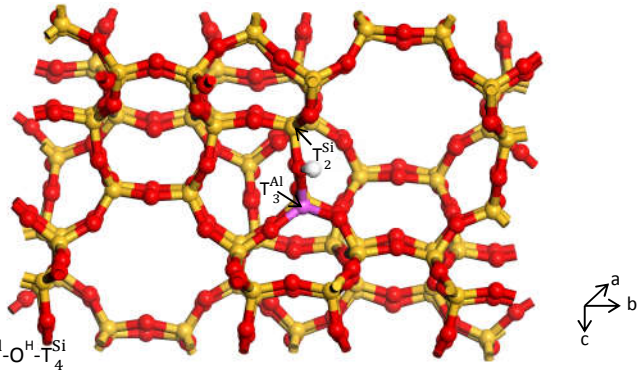


T^{Al}_2-O^H-T^{Si}_8

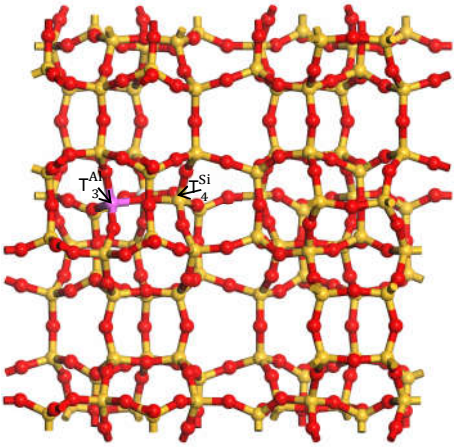




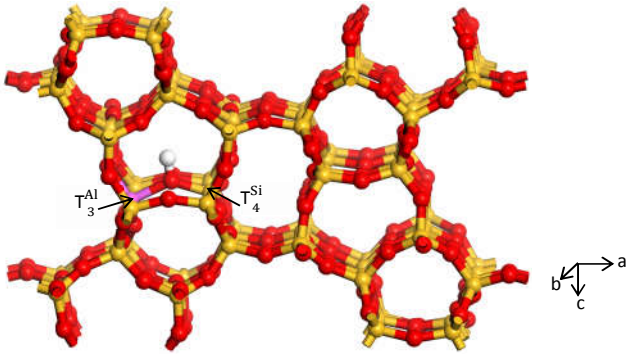
T^{Al}_{3}-O^{H}-T^{Si}_{2}



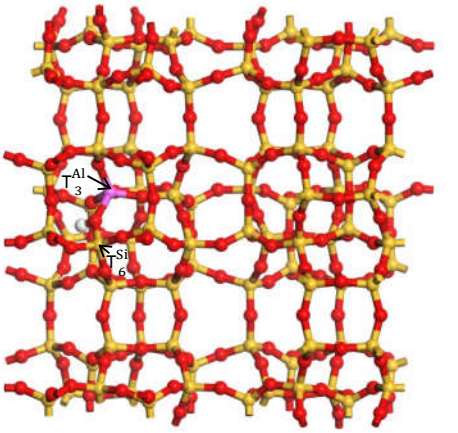
T^{Al}_{3}-O^{H}-T^{Si}_{4}



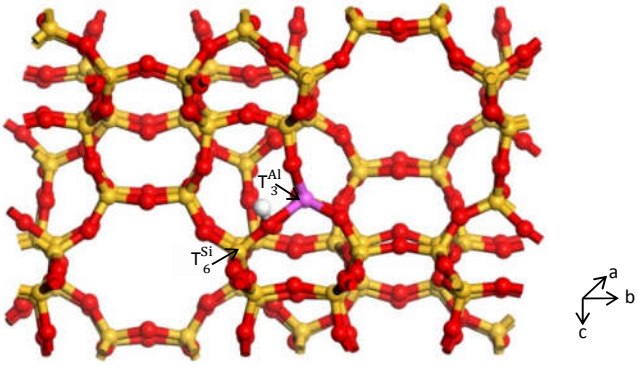
T^{Al}_{3}-O^{H}-T^{Si}_{4}



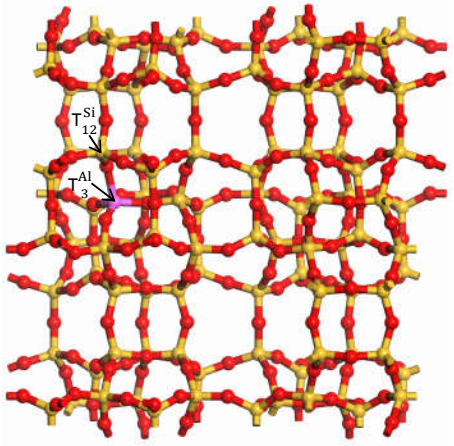
T^{Al}_{3}-O^{H}-T^{Si}_{6}



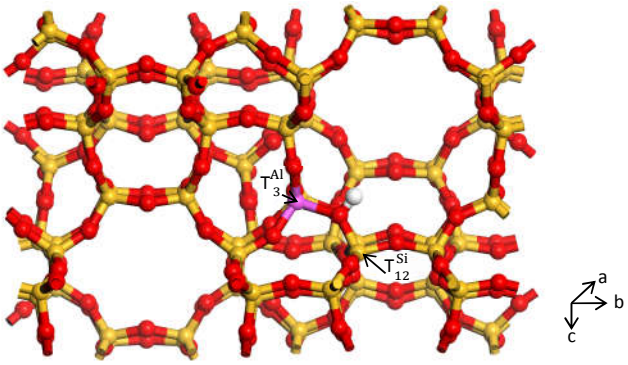
T^{Al}_{3}-O^{H}-T^{Si}_{6}

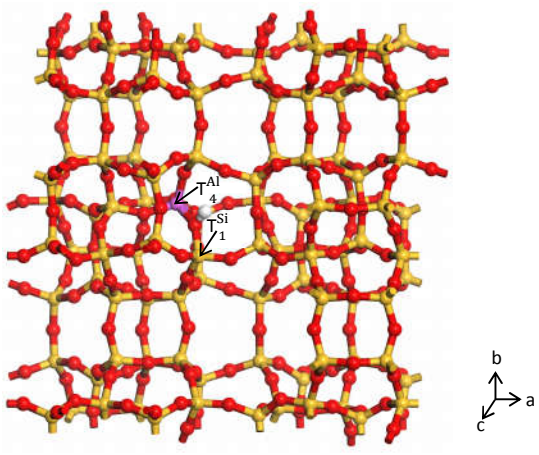


T^{Al}_{3}-O^{H}-T^{Si}_{12}

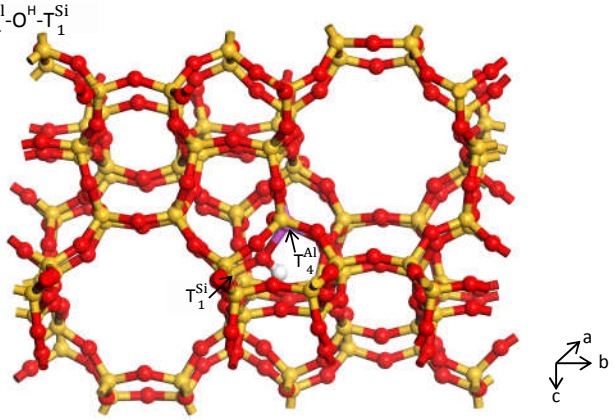


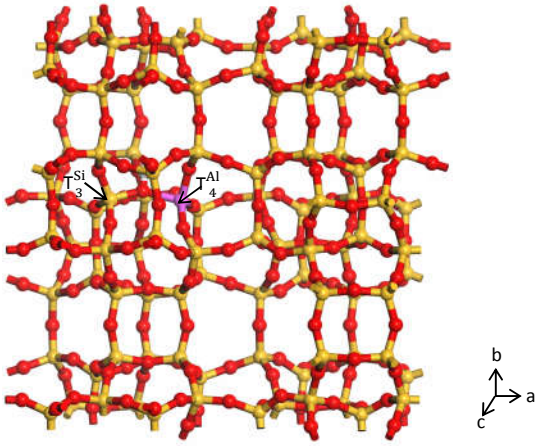
T^{Al}_{3}-O^{H}-T^{Si}_{12}



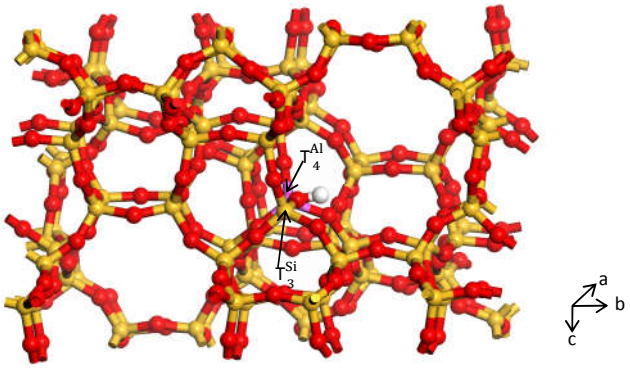


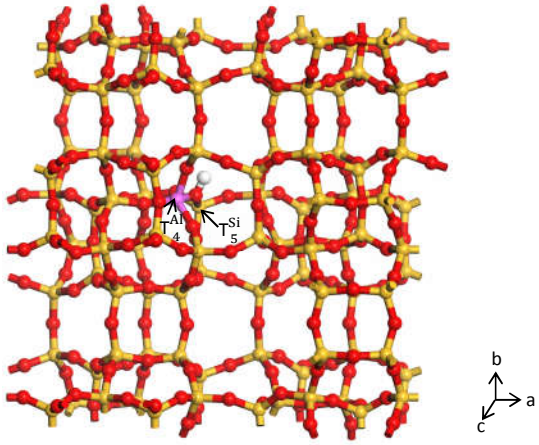
T^{Al}_4-O^H-T^{Si}_1



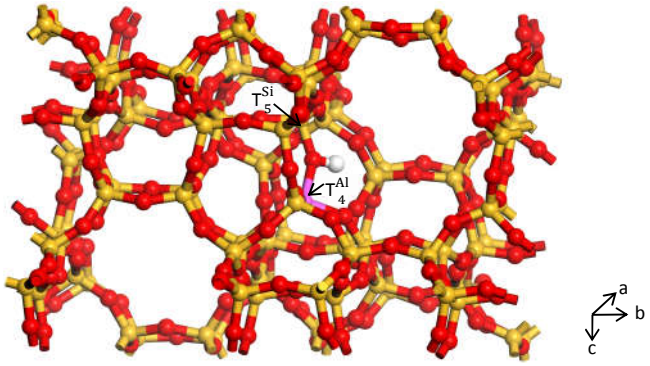


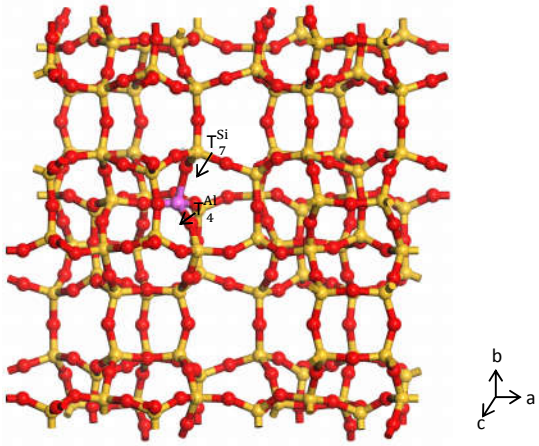
T^{Al}_4-O^H-T^{Si}_3



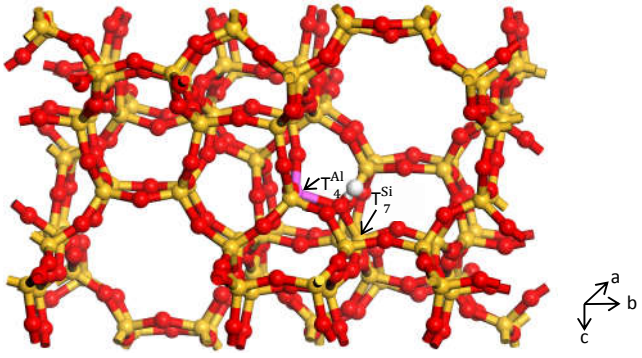


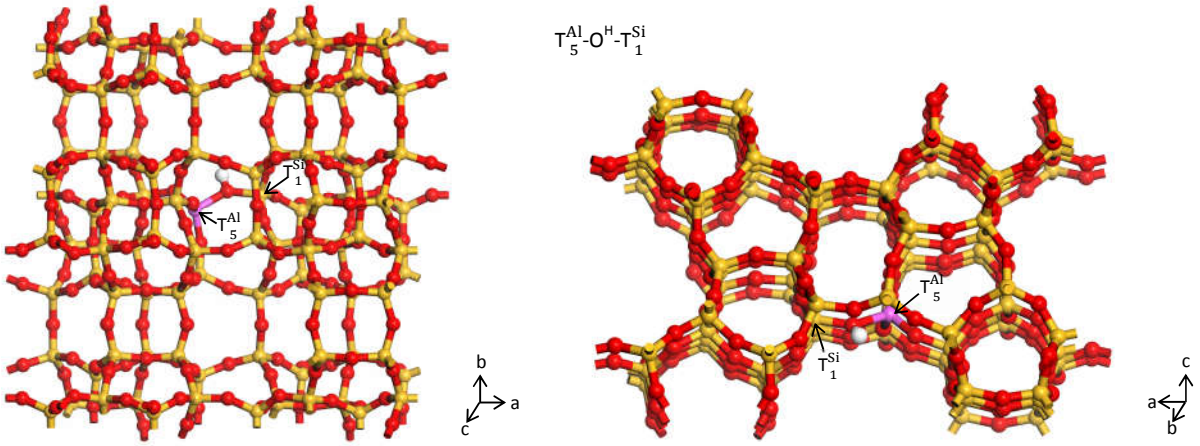
T^{Al}_4-O^H-T^{Si}_5

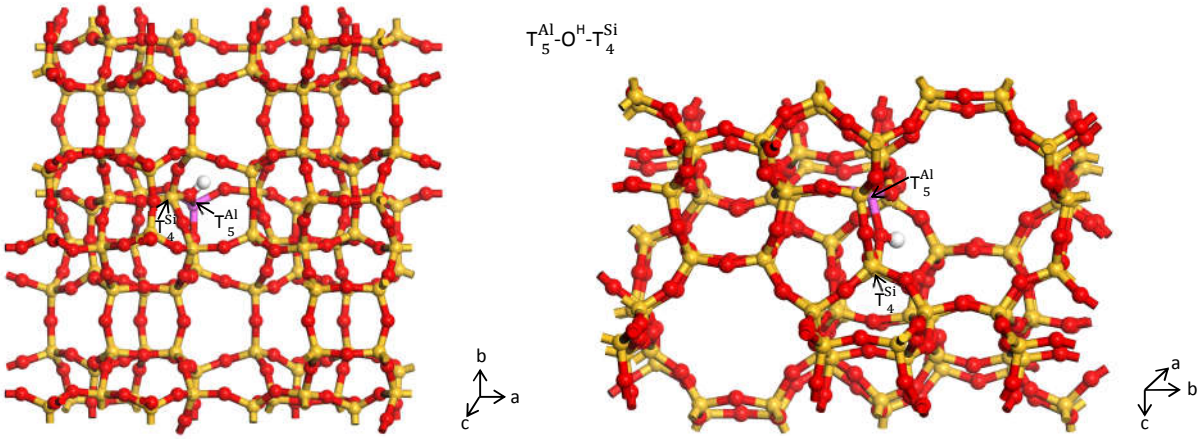


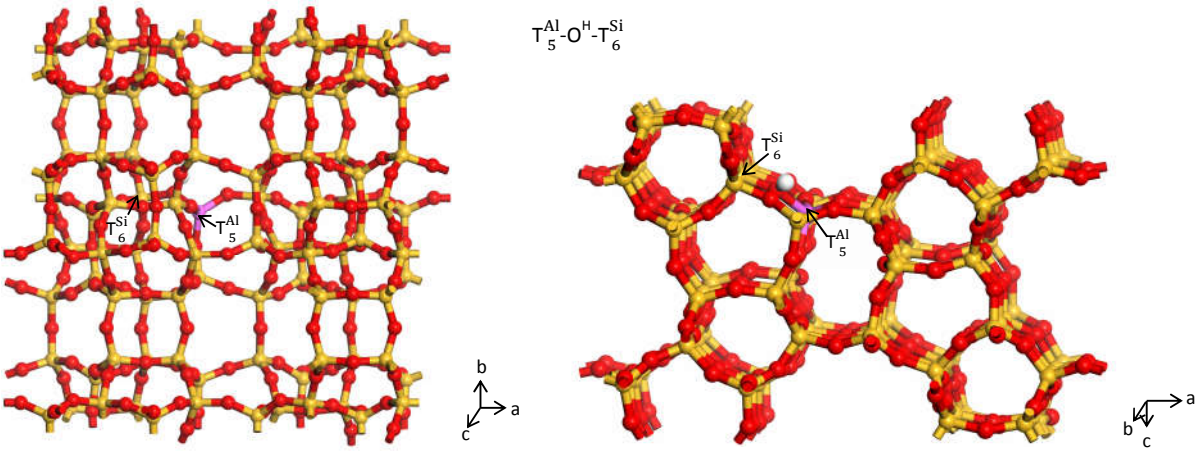


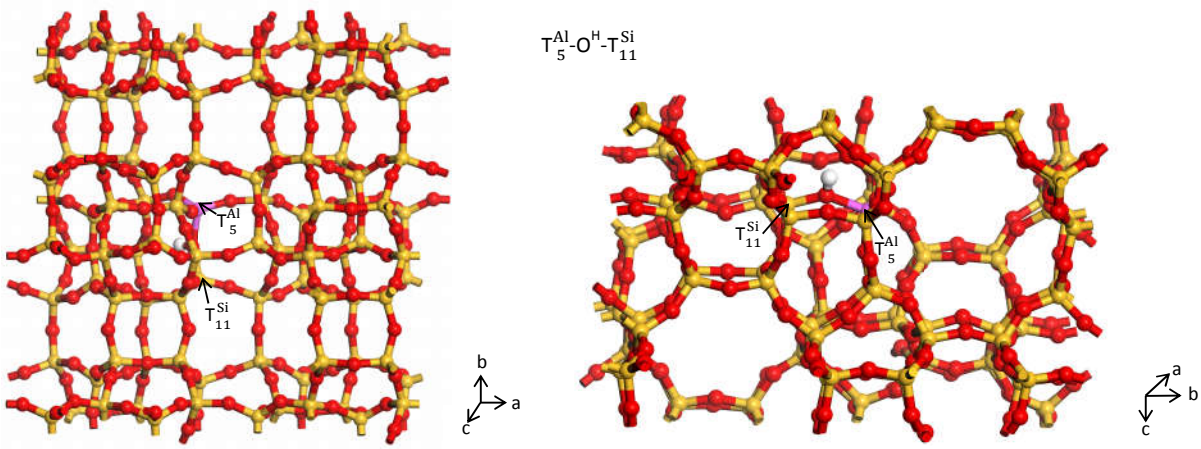
T^{Al}_4-O^H-T^{Si}_7

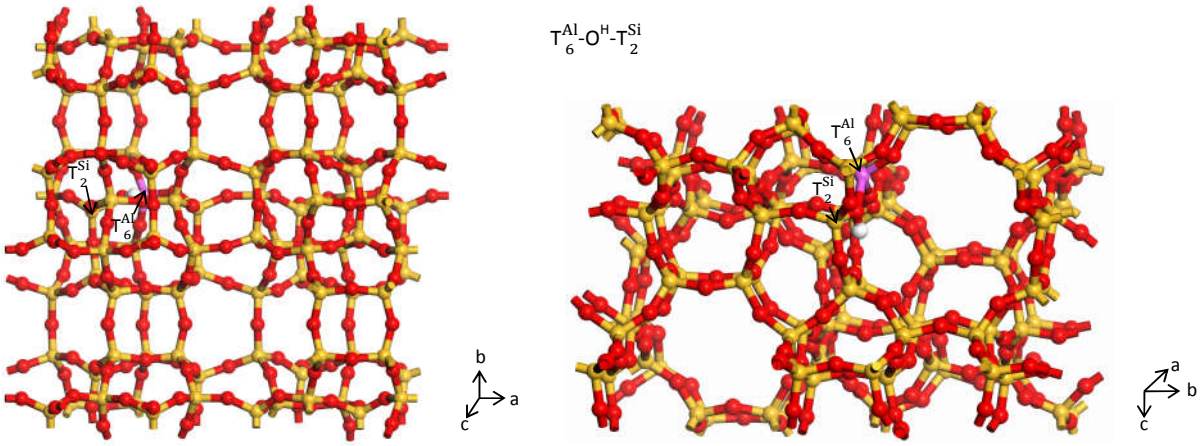


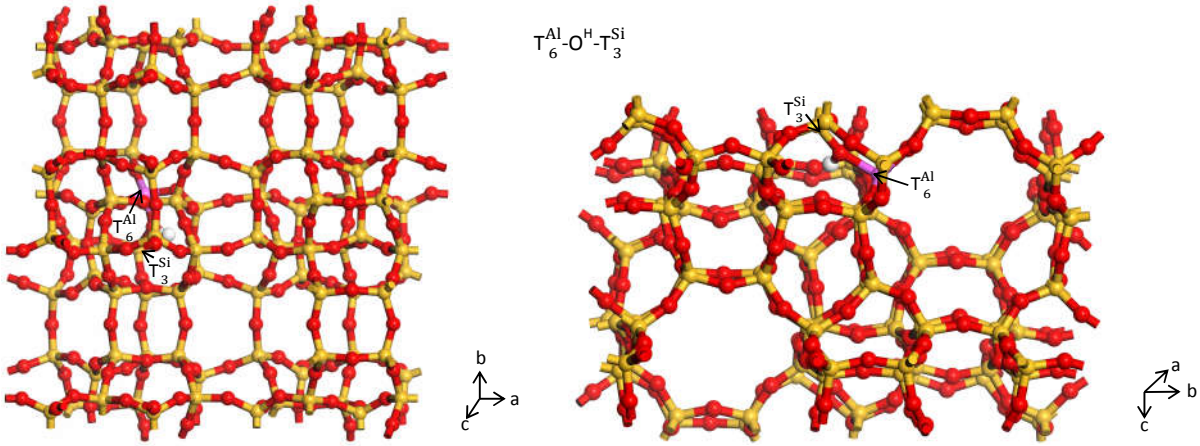

T_5^{Al}-O^H-T_1^{Si}
T^{Si}_1
T^{Al}_5
b
a
c
T^{Al}_5
T^{Si}_1
c
a
b

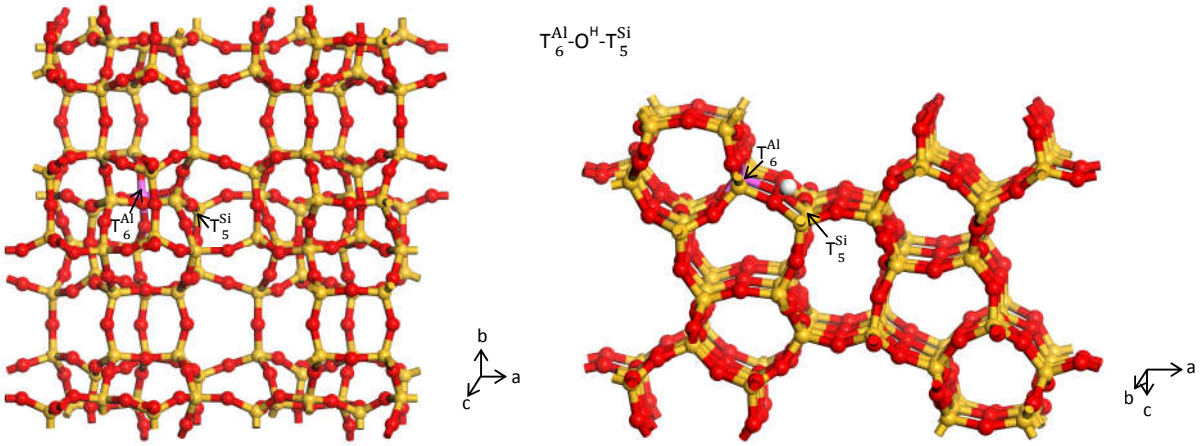

T_5^{Al}-O^H-T_4^{Si}
T^{Si}_4
T^{Al}_5
b
a
c
T^{Al}_5
T^{Si}_4
a
b
c

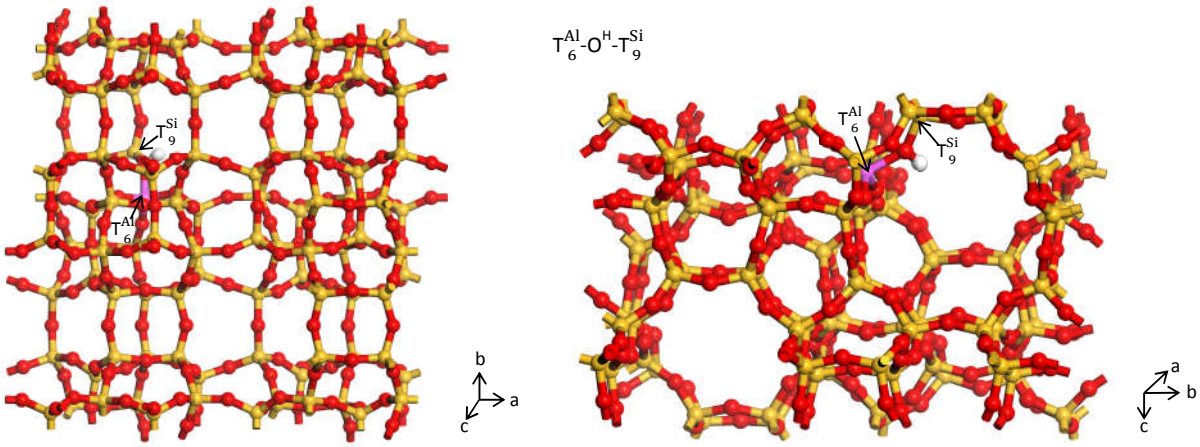

T_5^{Al}-O^H-T_6^{Si}
T^{Si}_6
T^{Al}_5
b
a
c
T^{Si}_6
T^{Al}_5
a
b
c

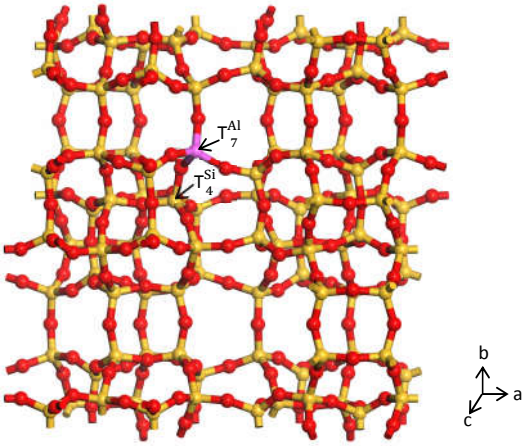

T_5^{Al}-O^H-T_{11}^{Si}
T^{Al}_5
T^{Si}_{11}
b
a
c
T^{Si}_{11}
T^{Al}_5
a
b
c


T^{Al}_6-O^H-T^{Si}_2
T^{Si}_2
T^{Al}_6
b
a
c
T^{Al}_6
T^{Si}_2
a
b
c

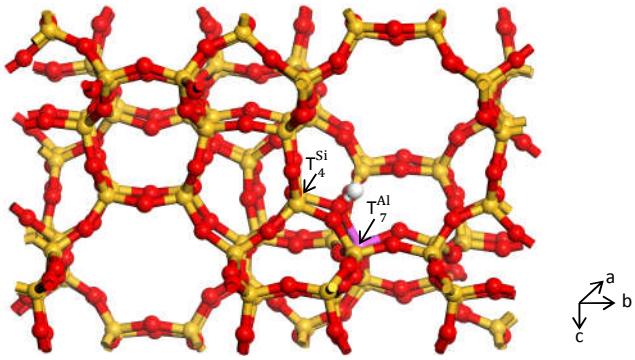

T^{Al}_6-O^H-T^{Si}_3
T^{Al}_6
T^{Si}_3
b
a
c
T^{Si}_3
T^{Al}_6
a
b
c

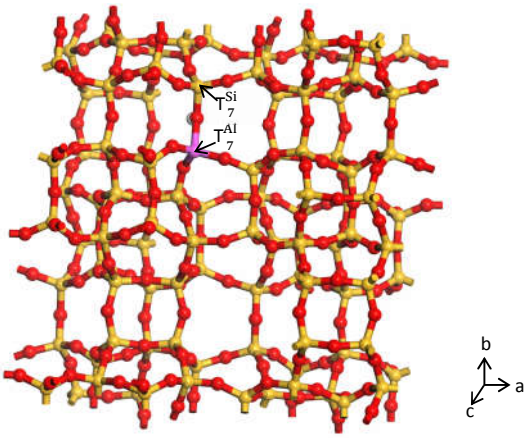

T^{Al}_6-O^H-T^{Si}_5
T^{Al}_6
T^{Si}_5
b
a
c
T^{Al}_6
T^{Si}_5
a
b
c


T^{Al}_6-O^H-T^{Si}_9
T^{Si}_9
T^{Al}_6
b
a
c
T^{Al}_6
T^{Si}_9
a
b
c

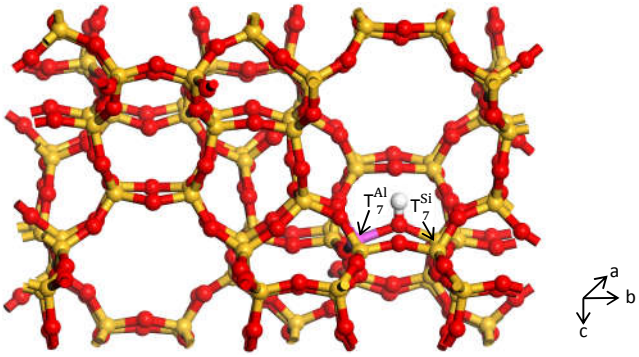


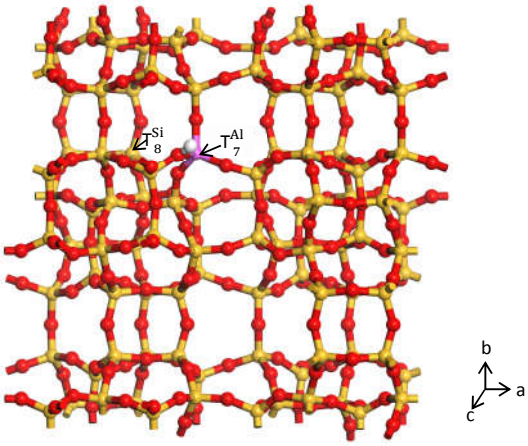
T^{Al}_{7}-O^{H}-T^{Si}_{4}



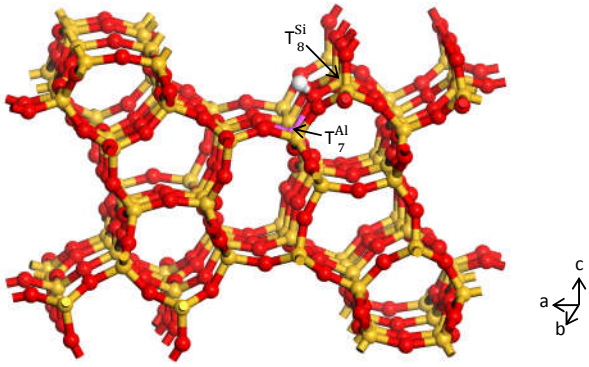


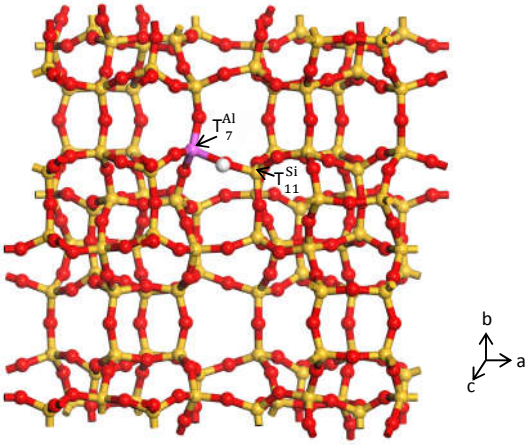
T^{Al}_{7}-O^{H}-T^{Si}_{7}



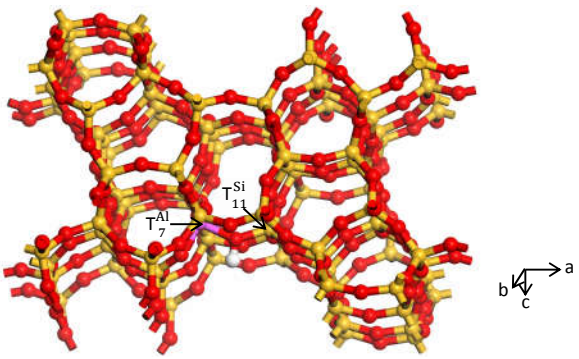


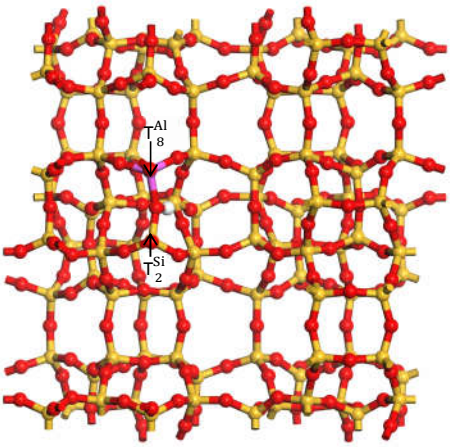
T^{Al}_{7}-O^{H}-T^{Si}_{8}



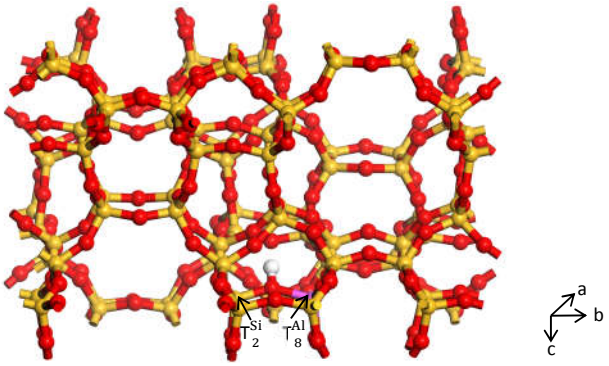


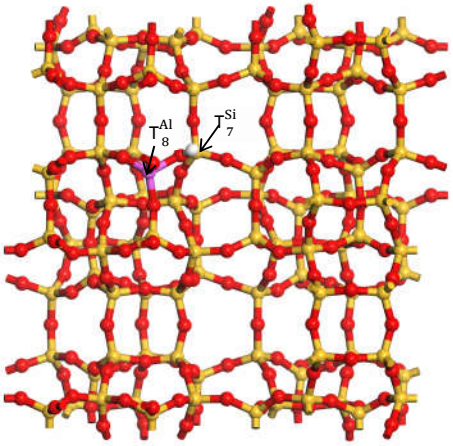
T^{Al}_{7}-O^{H}-T^{Si}_{11}



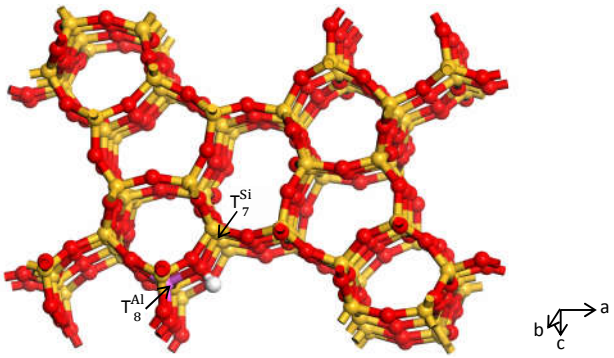


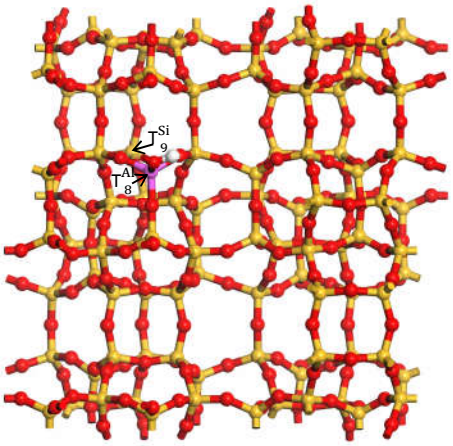
T_8^{Al}-O^H-T_2^{Si}



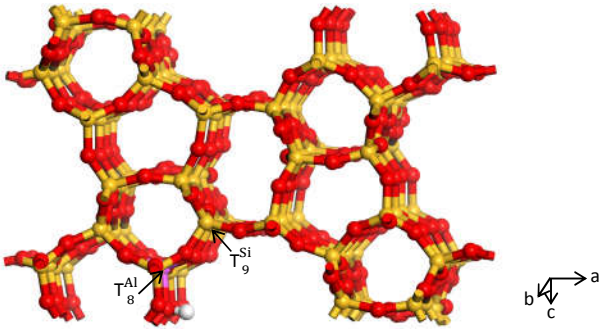


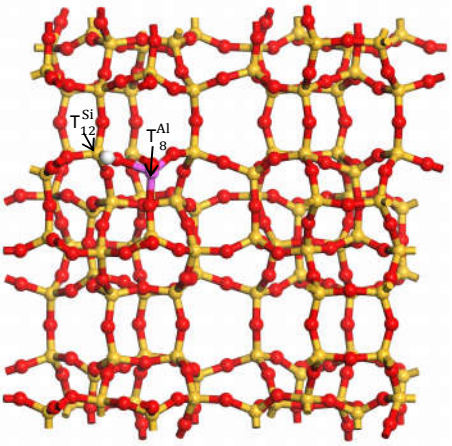
T_8^{Al}-O^H-T_7^{Si}



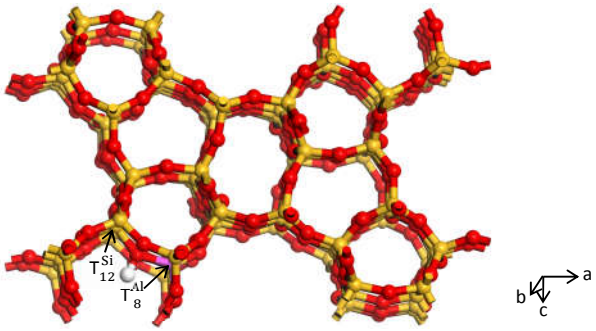


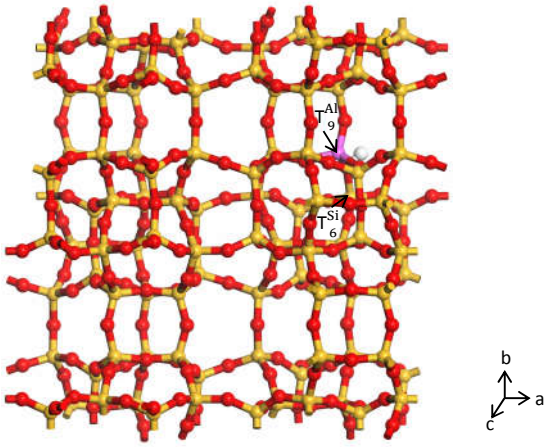
T_8^{Al}-O^H-T_9^{Si}



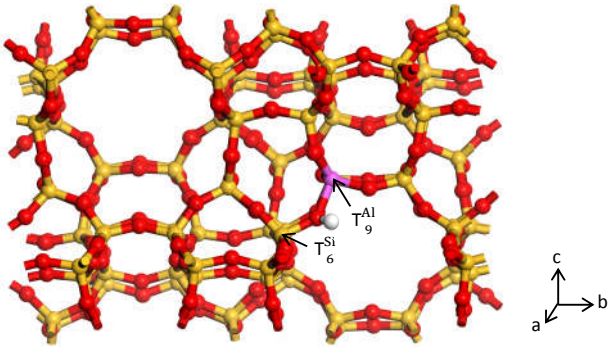


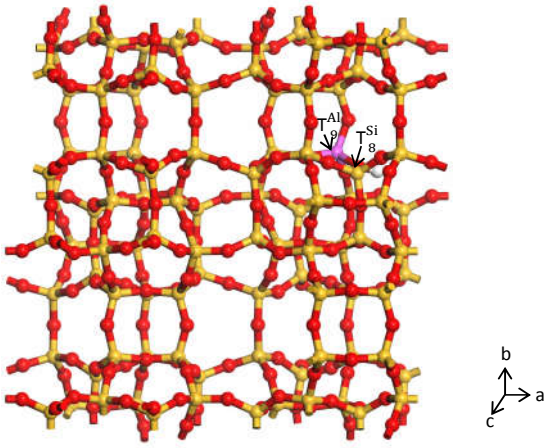
T_8^{Al}-O^H-T_{12}^{Si}



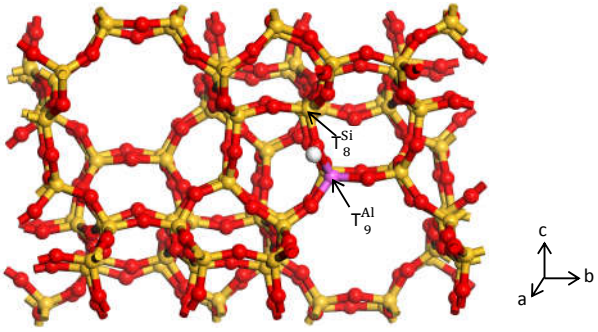


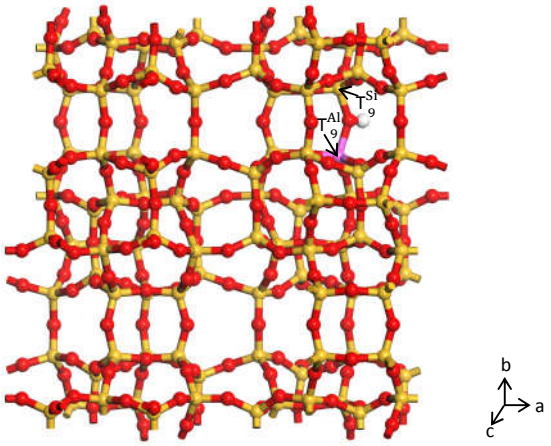
T^{Al}_{9}-O^{H}-T^{Si}_{6}



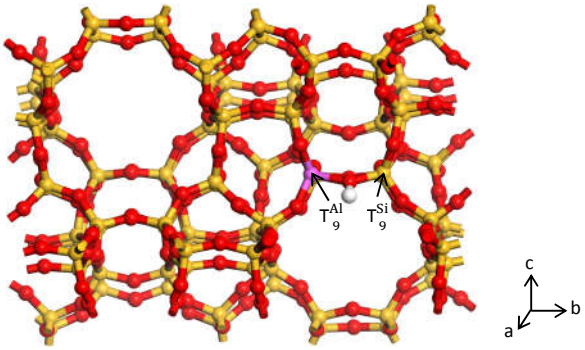


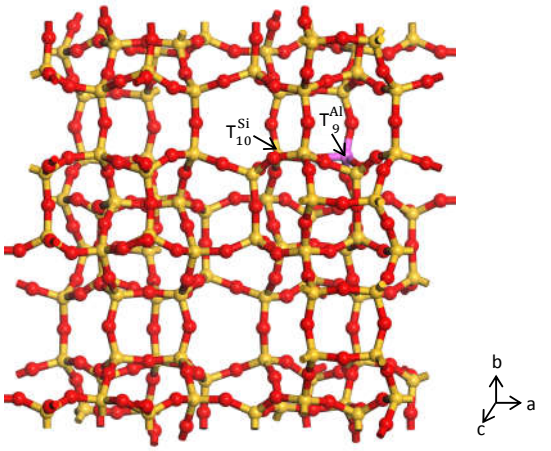
T^{Al}_{9}-O^{H}-T^{Si}_{8}



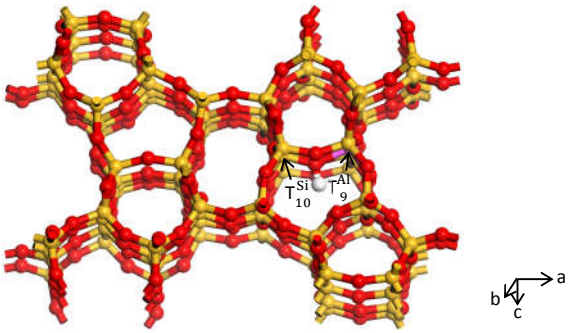


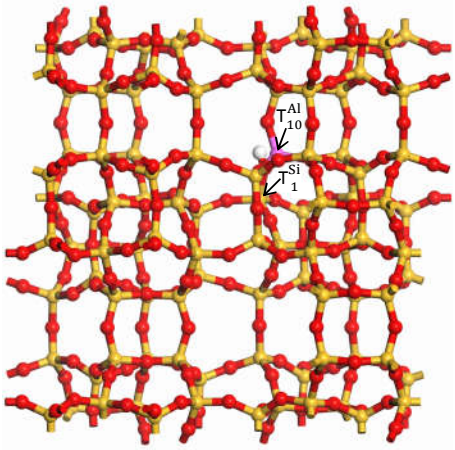
T^{Al}_{9}-O^{H}-T^{Si}_{9}



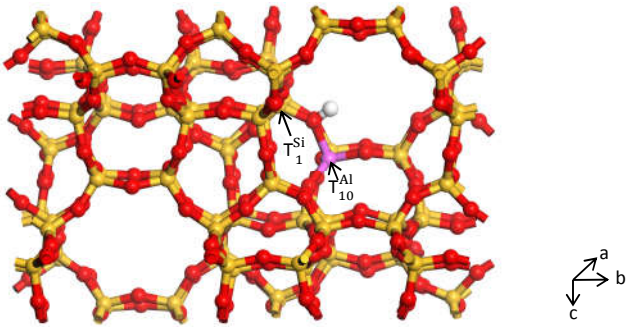


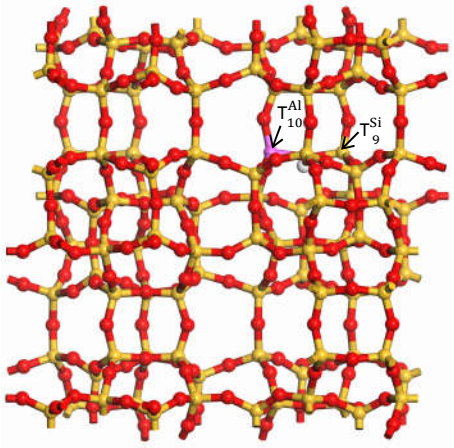
T^{Al}_{9}-O^{H}-T^{Si}_{10}



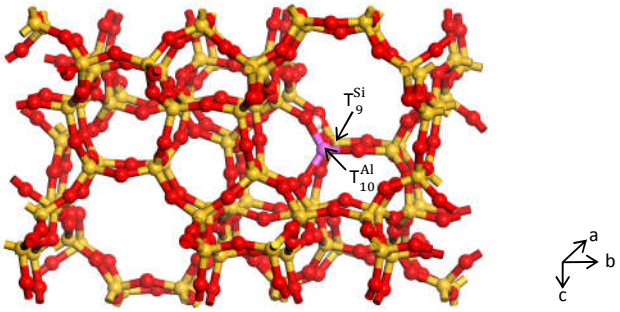


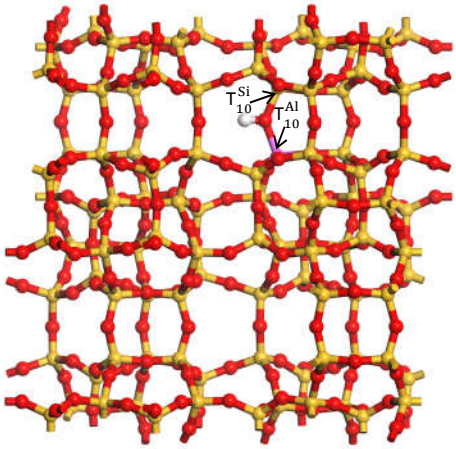
T^{Al}_{10}-O^{H}-T^{Si}_{1}



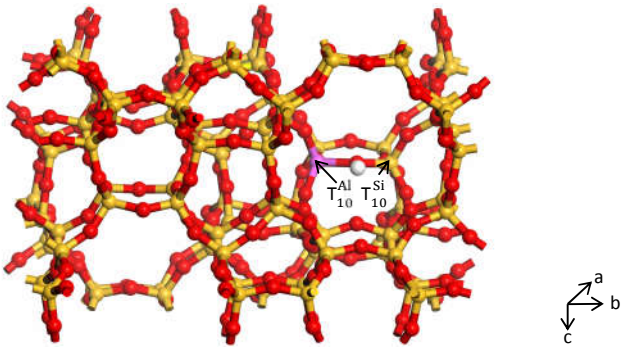


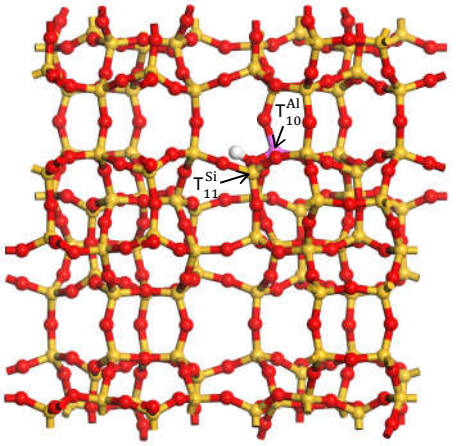
T^{Al}_{10}-O^{H}-T^{Si}_{9}



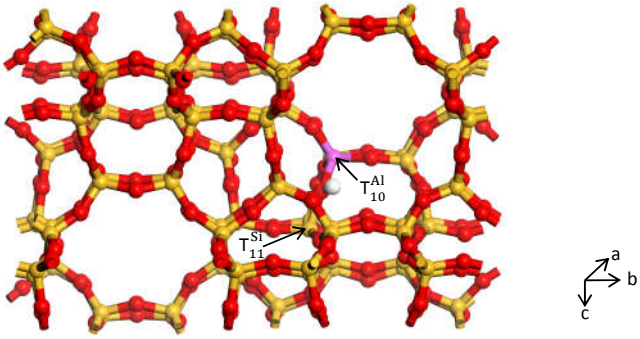


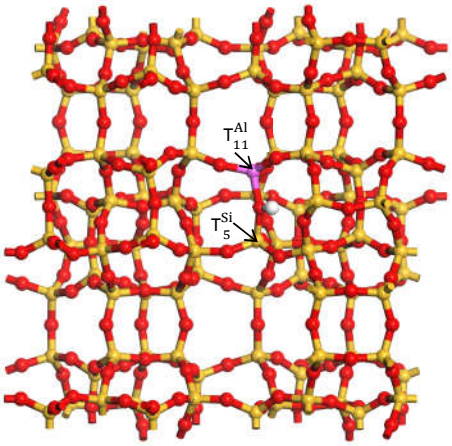
T^{Al}_{10}-O^{H}-T^{Si}_{10}



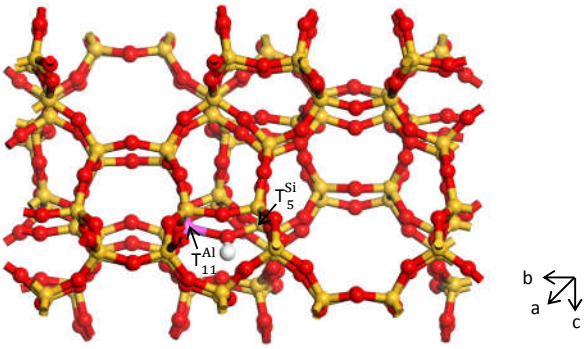


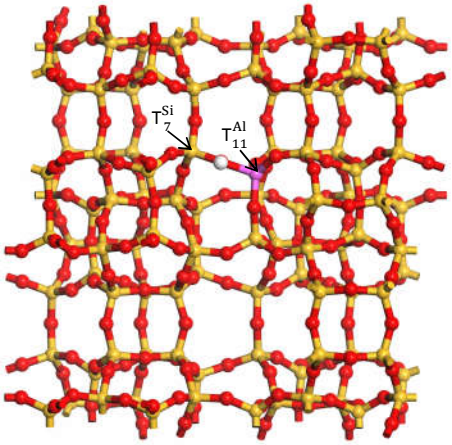
T^{Al}_{10}-O^{H}-T^{Si}_{11}



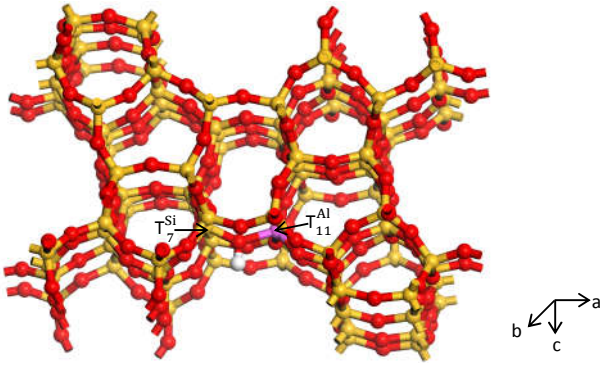


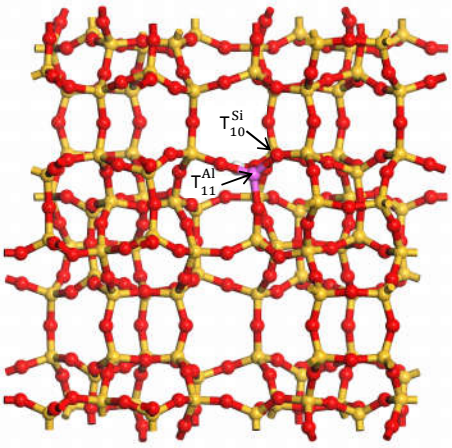
T^{Al}_{11}-O^{H}-T^{Si}_{5}



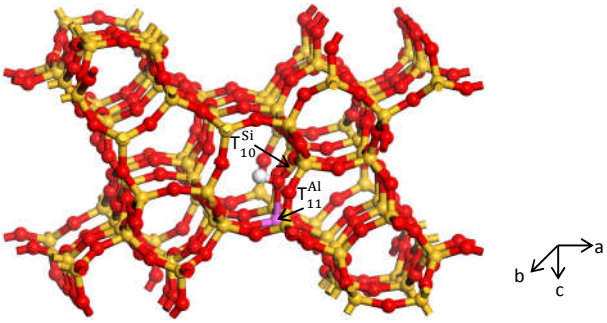


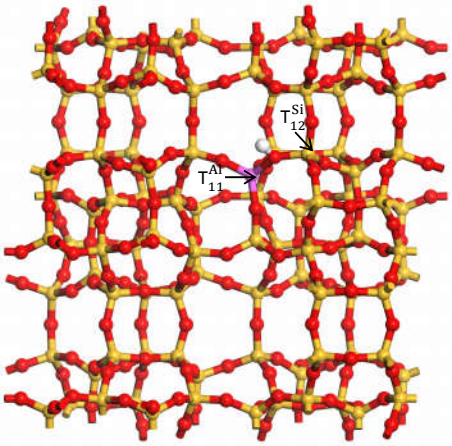
T^{Al}_{11}-O^{H}-T^{Si}_{7}



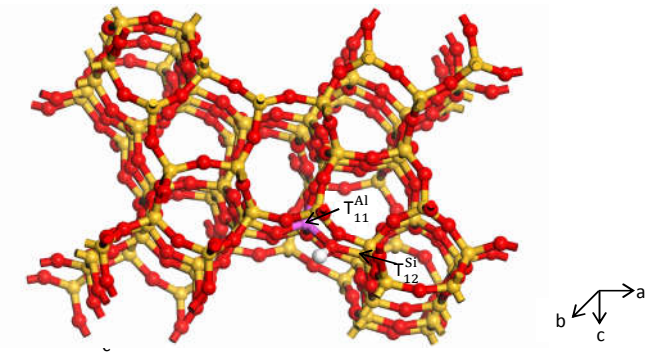


T^{Al}_{11}-O^{H}-T^{Si}_{10}





T^{Al}_{11}-O^{H}-T^{Si}_{12}



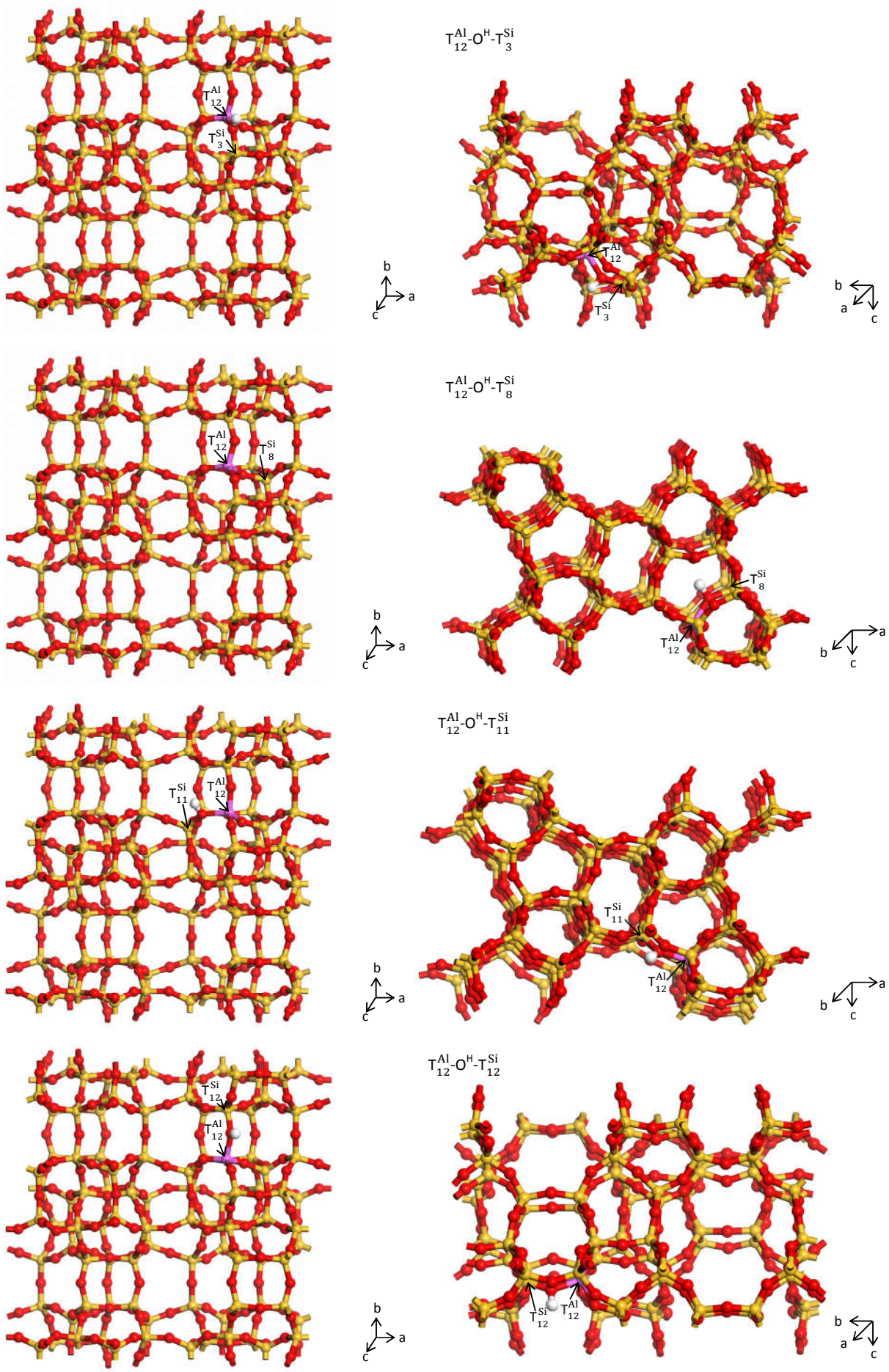


Figure S1 The optimized configurations of all 48 acidic sites for one Al substitution in the framework

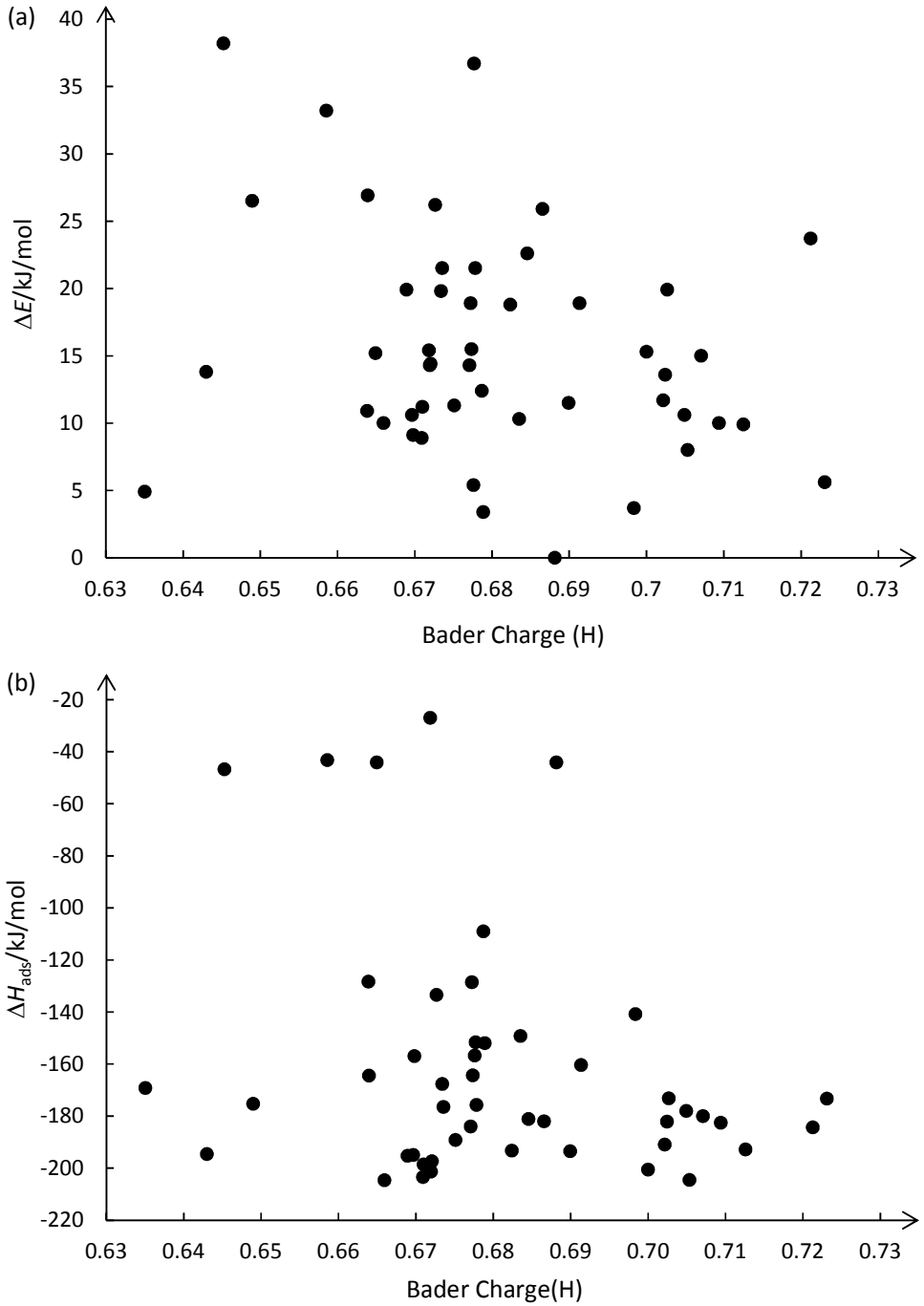
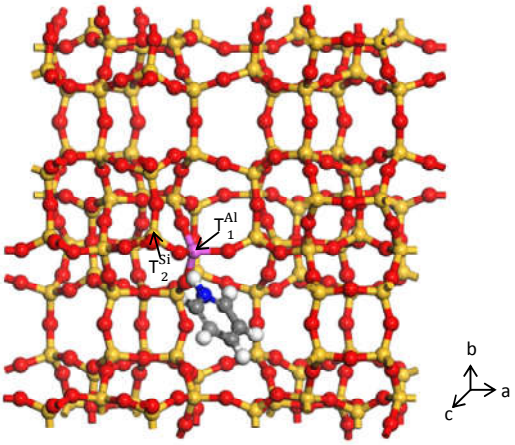
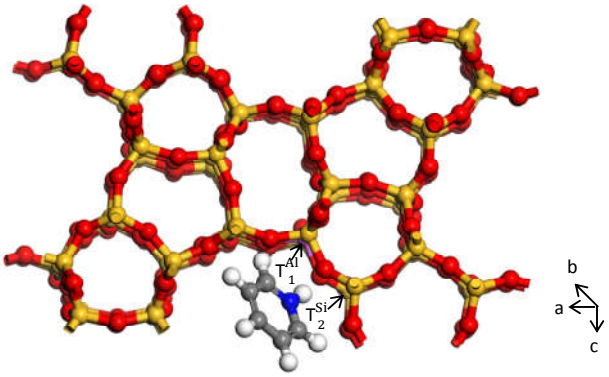
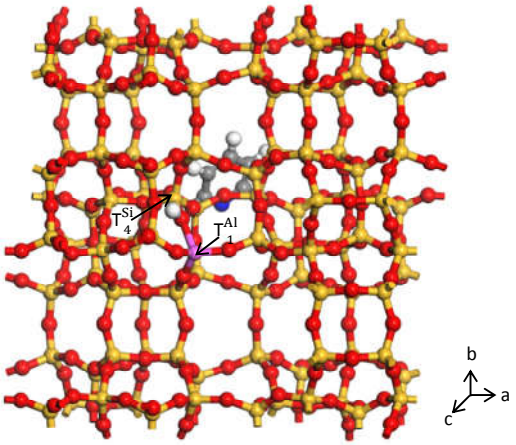


Figure S2 Calculated Bader charge as a function of (a) relative energy ΔE of all 48 acid sites and (b) pyridine adsorption enthalpy ΔH_{ads}

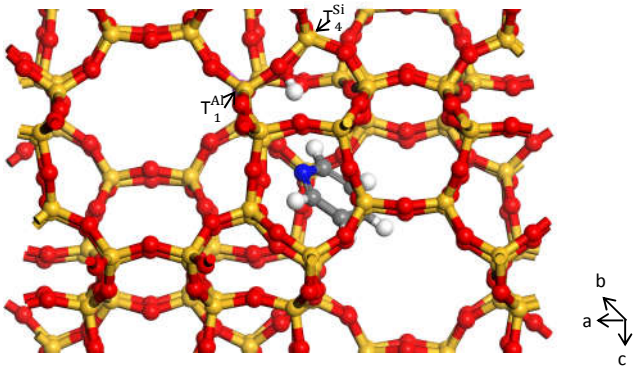


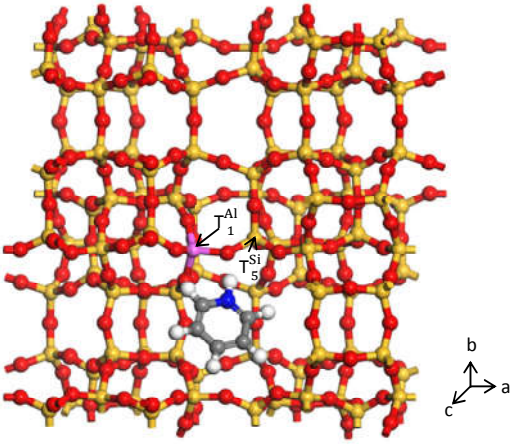
T^{Al}_1-O^H-T^{Si}_2



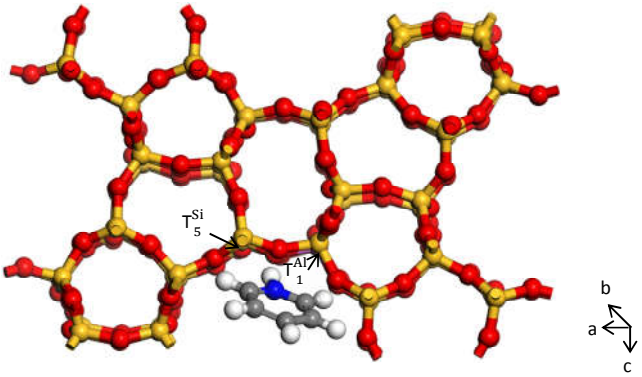


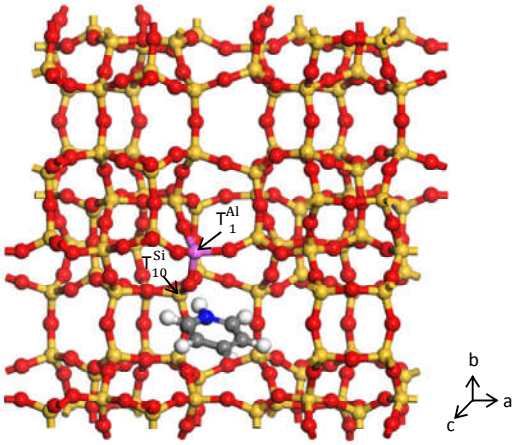
T^{Al}_1-O^H-T^{Si}_4



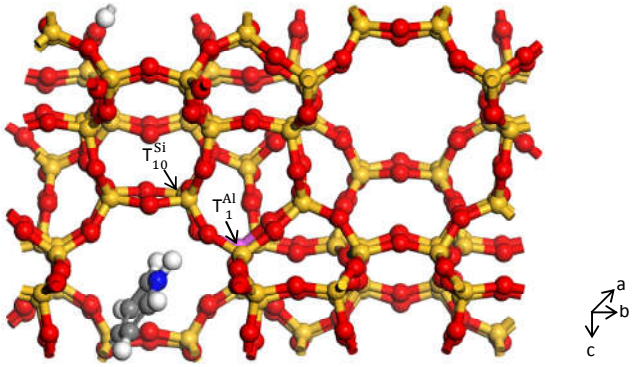


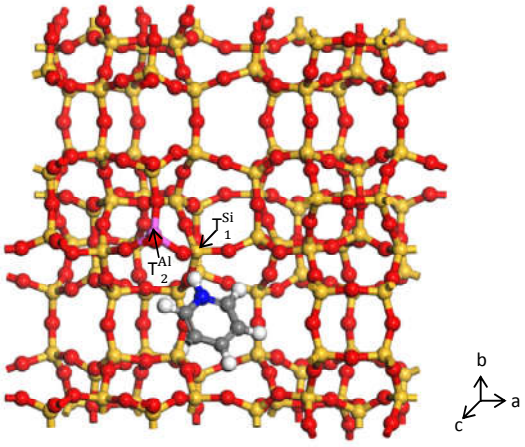
T^{Al}_1-O^H-T^{Si}_5



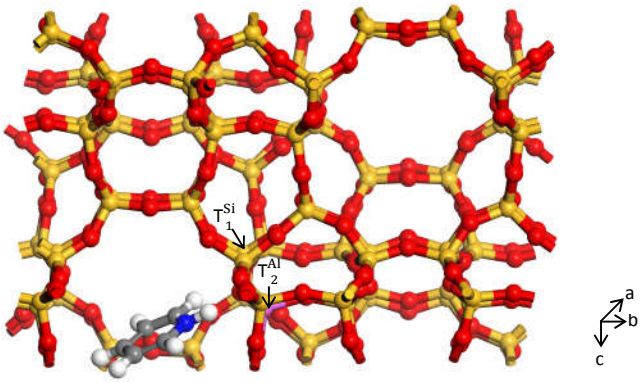


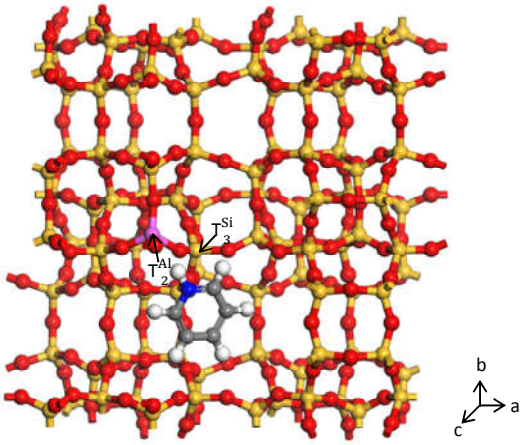
T^{Al}_1-O^H-T^{Si}_{10}



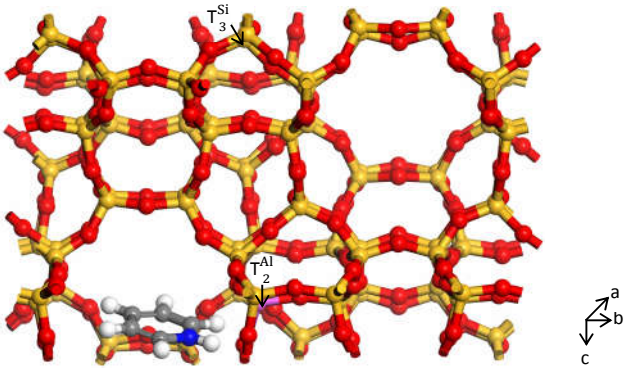


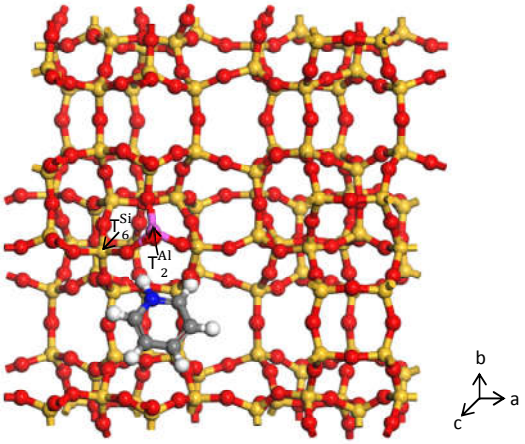
T^{Al}_{2}-O^{H}-T^{Si}_{1}



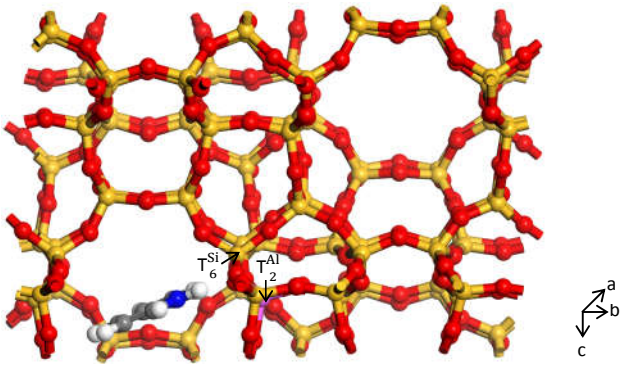


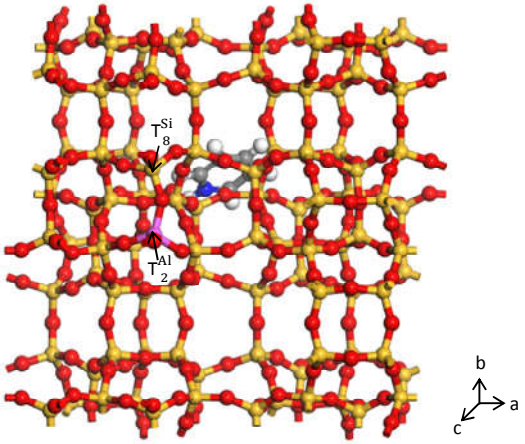
T^{Al}_{2}-O^{H}-T^{Si}_{3}



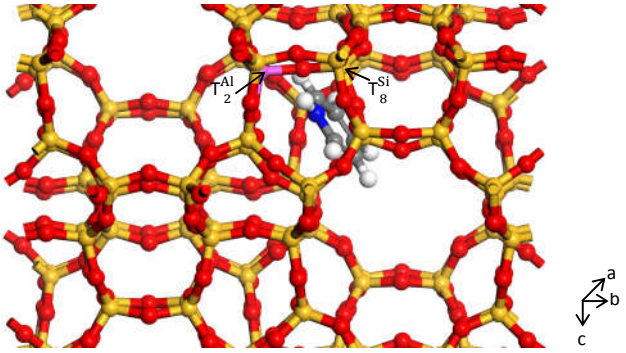


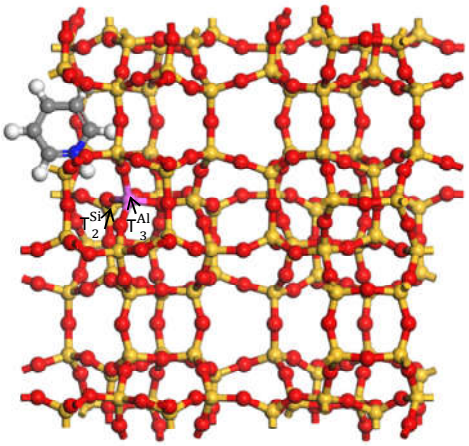
T^{Al}_{2}-O^{H}-T^{Si}_{6}



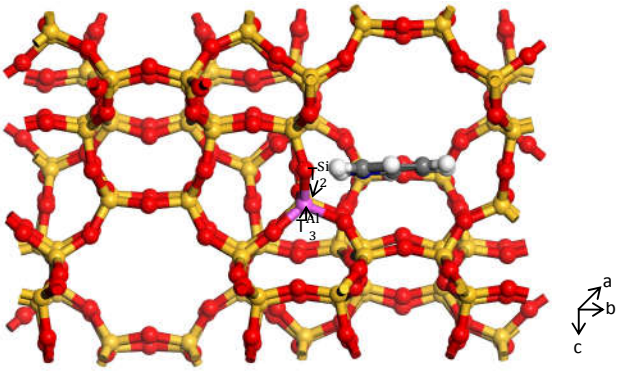


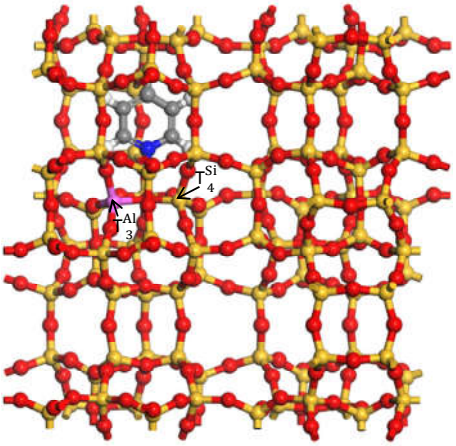
T^{Al}_{2}-O^{H}-T^{Si}_{8}



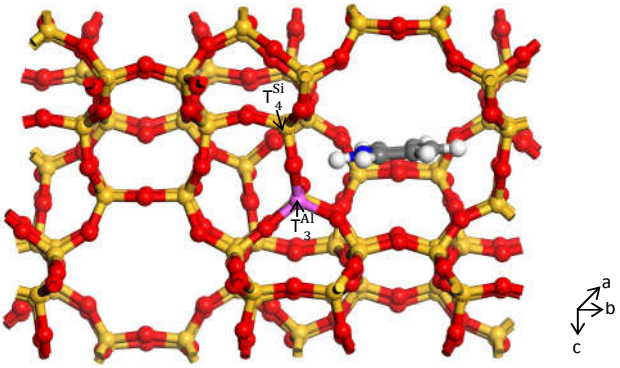


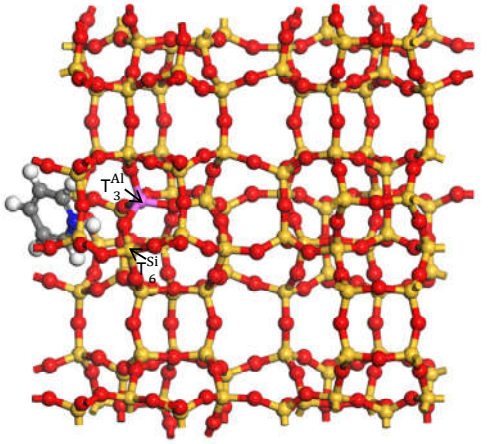
T_3^{Al}-O^H-T_2^{Si}



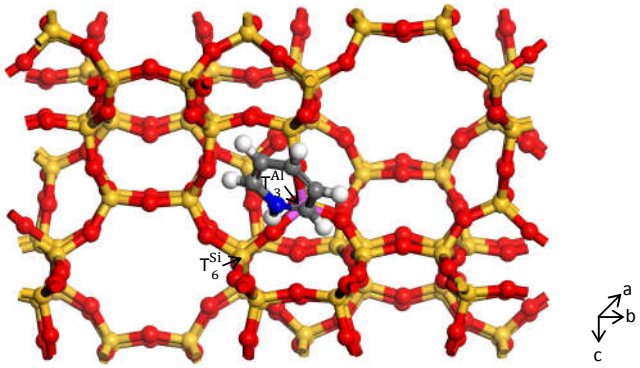


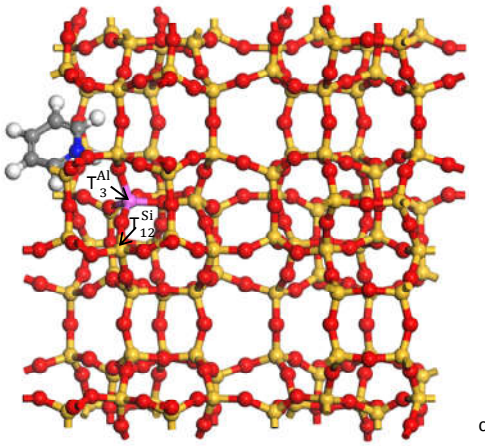
T_3^{Al}-O^H-T_4^{Si}



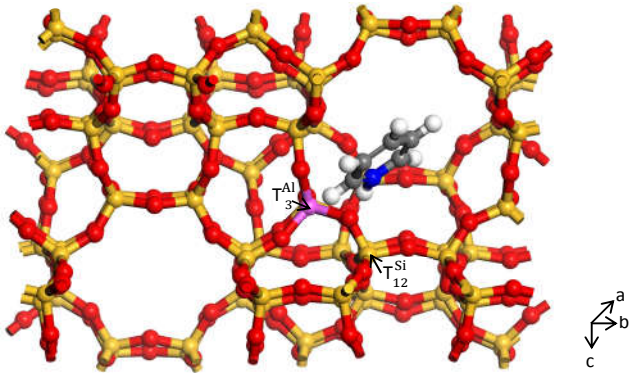


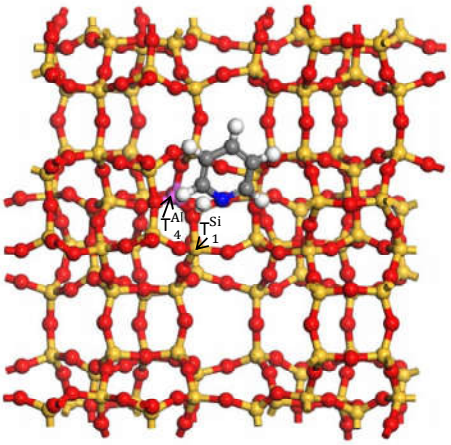
T_3^{Al}-O^H-T_6^{Si}



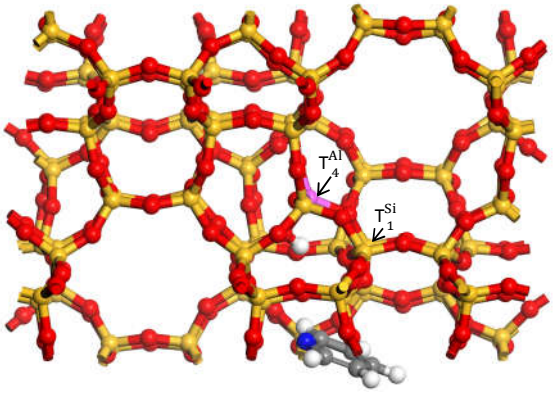


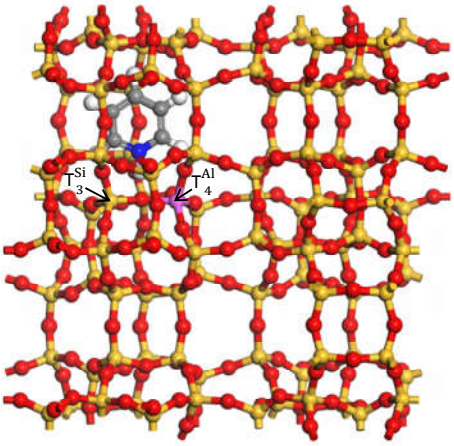
T_3^{Al}-O^H-T_{12}^{Si}



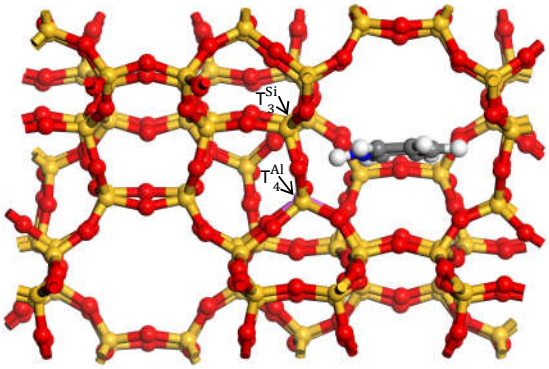


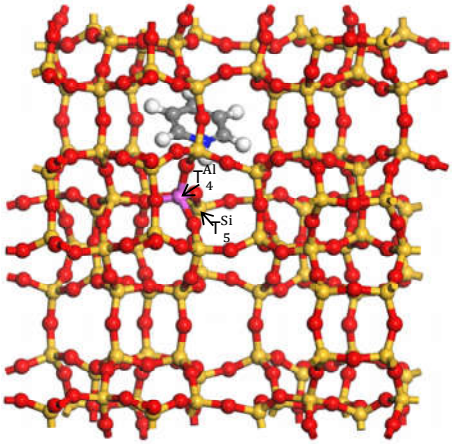
T_4^{Al}-O^H-T_1^{Si}



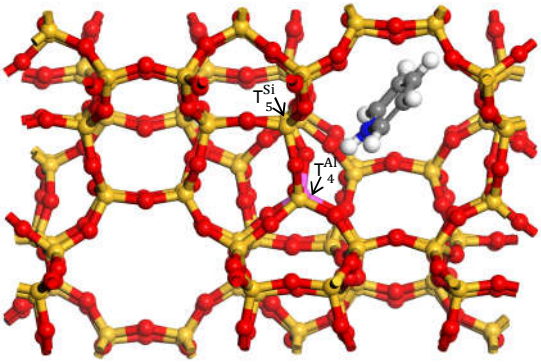


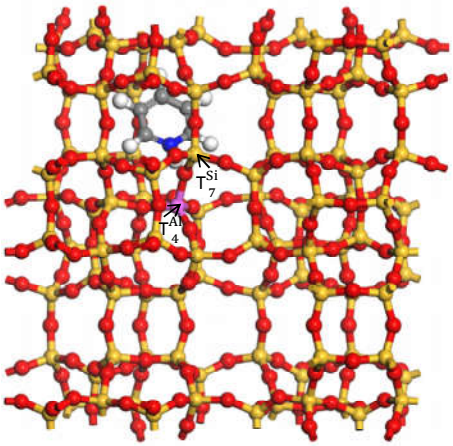
T_4^{Al}-O^H-T_3^{Si}



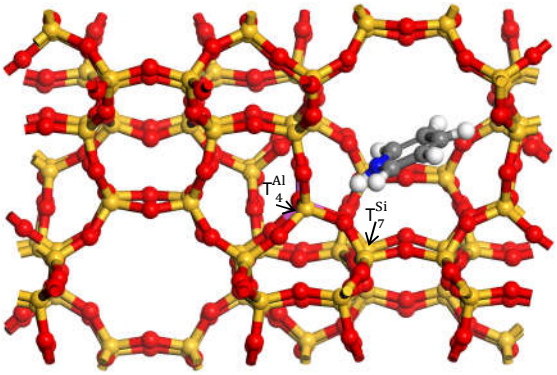


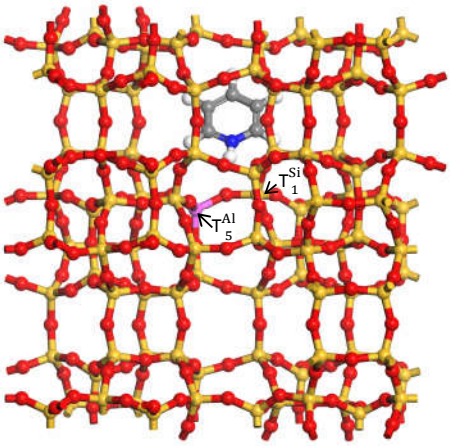
T_4^{Al}-O^H-T_5^{Si}



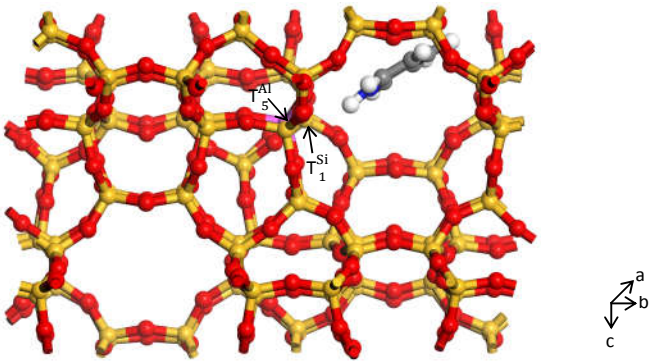


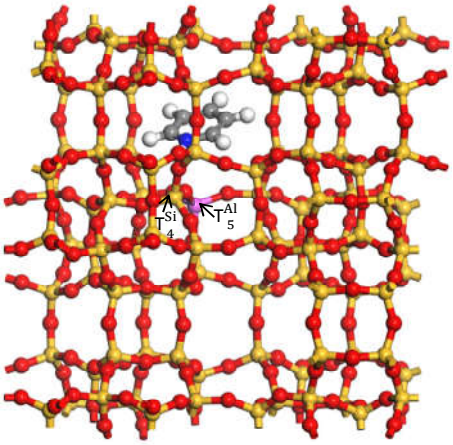
T_4^{Al}-O^H-T_7^{Si}



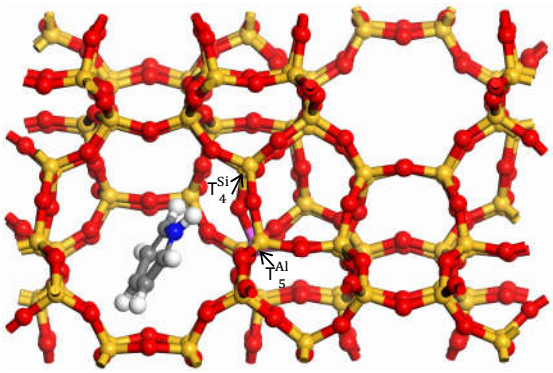


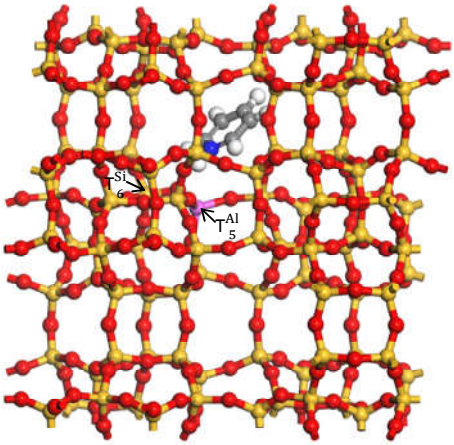
T_5^{Al}-O^H-T_1^{Si}



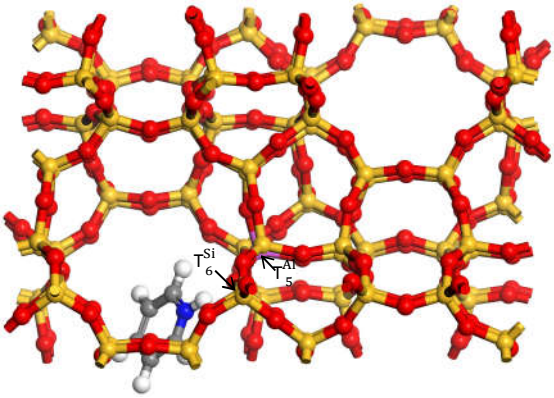


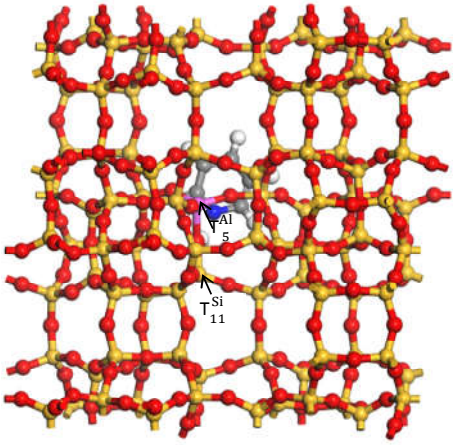
T_5^{Al}-O^H-T_4^{Si}



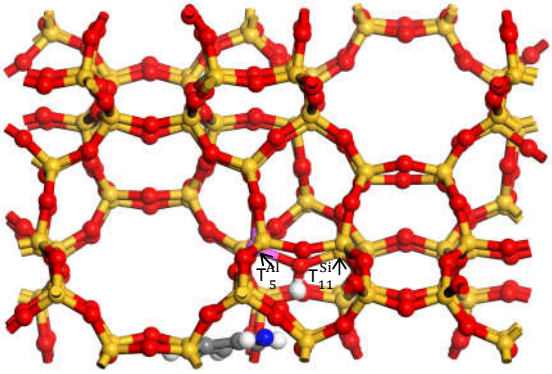


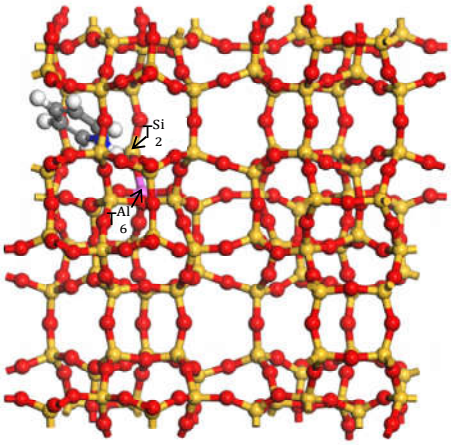
T_5^{Al}-O^H-T_6^{Si}



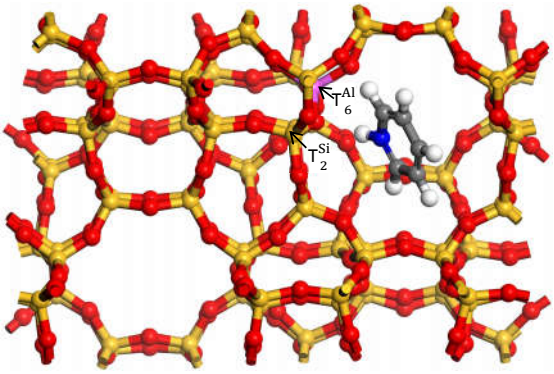


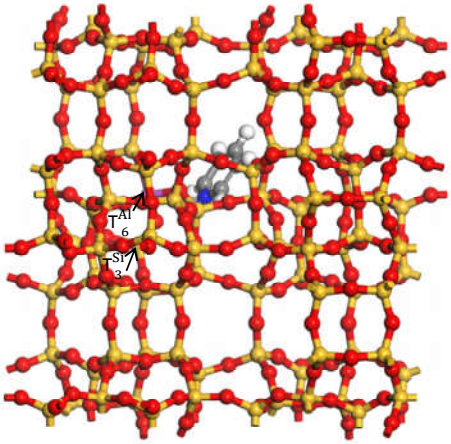
T_5^{Al}-O^H-T_{11}^{Si}



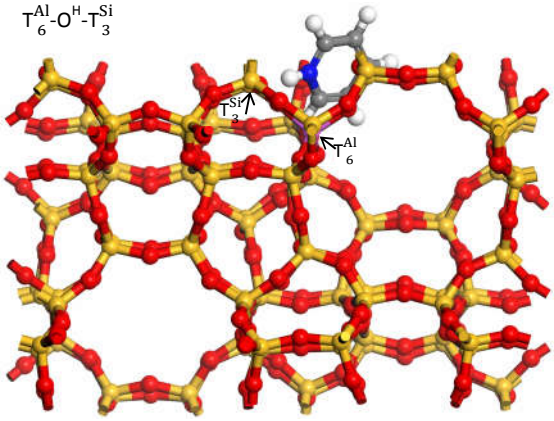


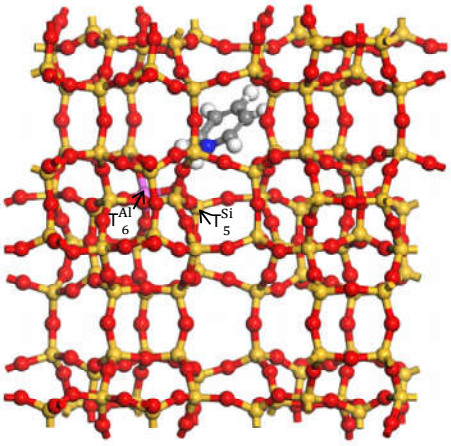
T^{Al}_{6}-O^{H}-T^{Si}_{2}



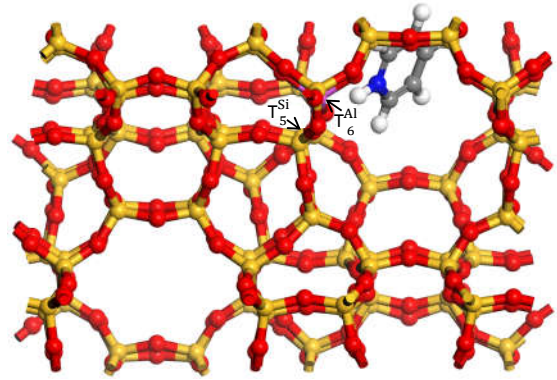


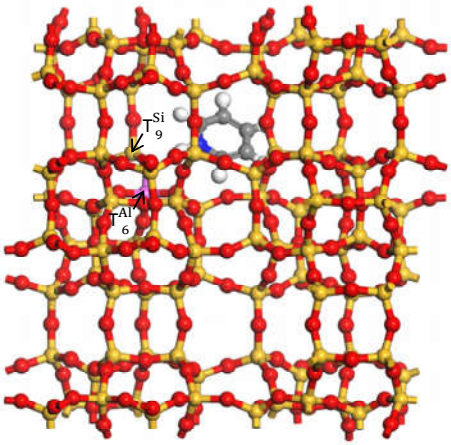
T^{Al}_{6}-O^{H}-T^{Si}_{3}



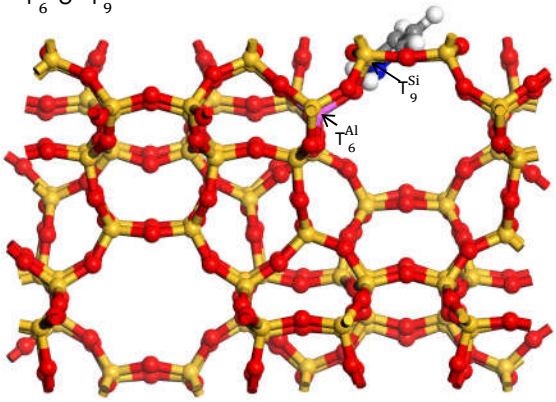


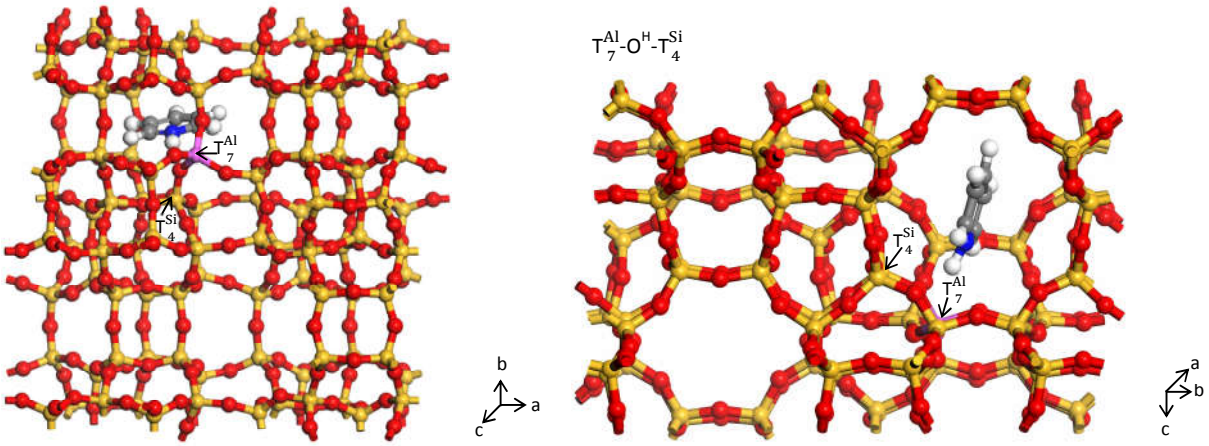
T^{Al}_{6}-O^{H}-T^{Si}_{5}

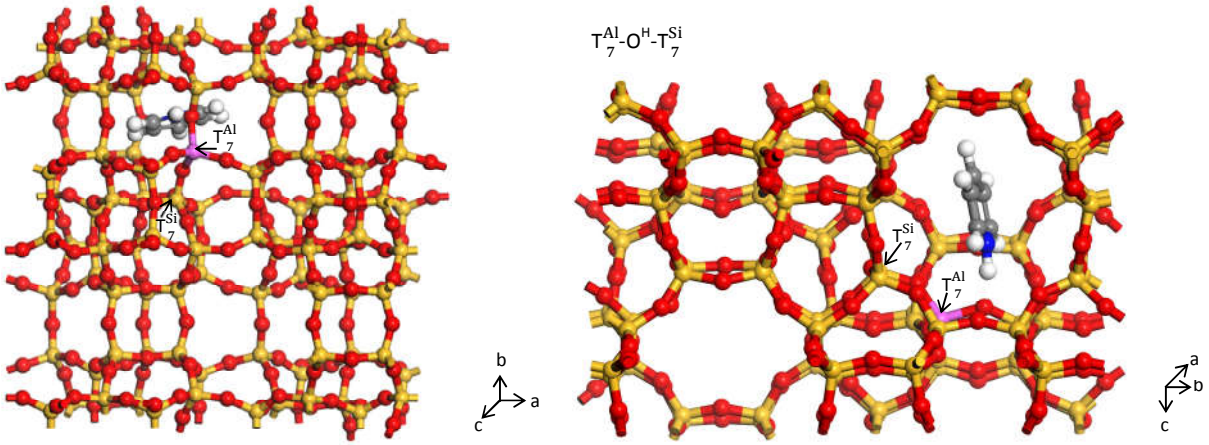


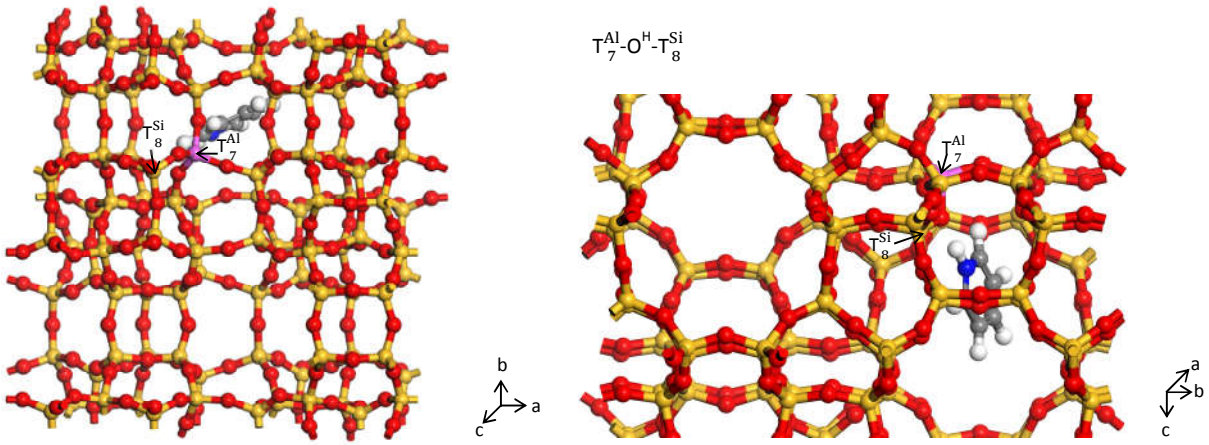


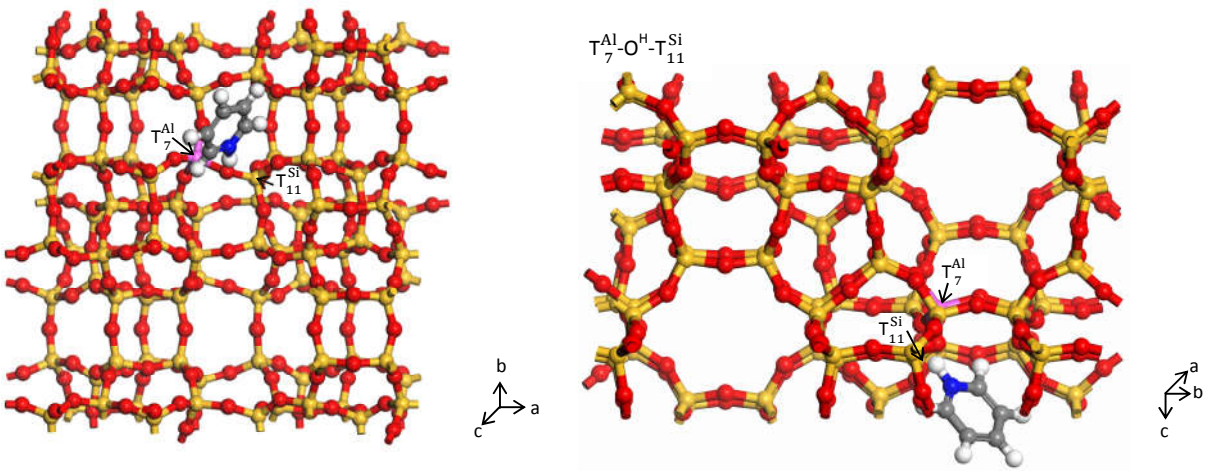
T^{Al}_{6}-O^{H}-T^{Si}_{9}

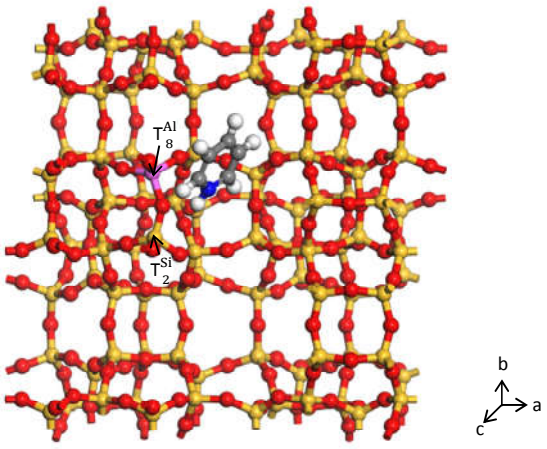



T^{Al}_{7}-O^{H}-T^{Si}_{4}
T^{Al}_{7}
T^{Si}_{4}
b
a
c
a
b
c

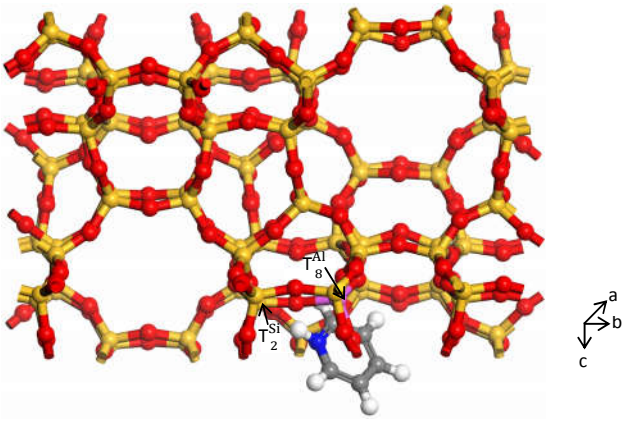

T^{Al}_{7}-O^{H}-T^{Si}_{7}
T^{Al}_{7}
T^{Si}_{7}
b
a
c
a
b
c

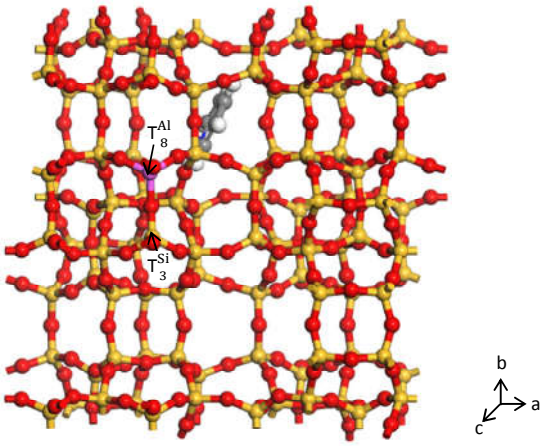

T^{Al}_{7}-O^{H}-T^{Si}_{8}
T^{Si}_{8}
T^{Al}_{7}
b
a
c
a
b
c


T^{Al}_{7}-O^{H}-T^{Si}_{11}
T^{Al}_{7}
T^{Si}_{11}
b
a
c
a
b
c

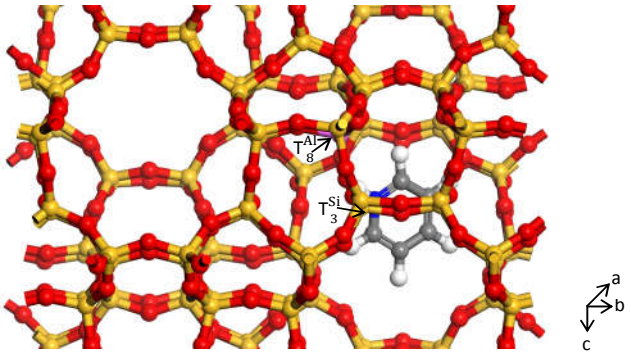


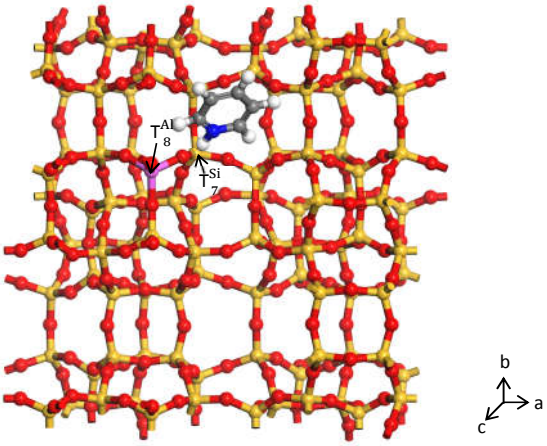
T_8^{Al}-O^H-T_2^{Si}



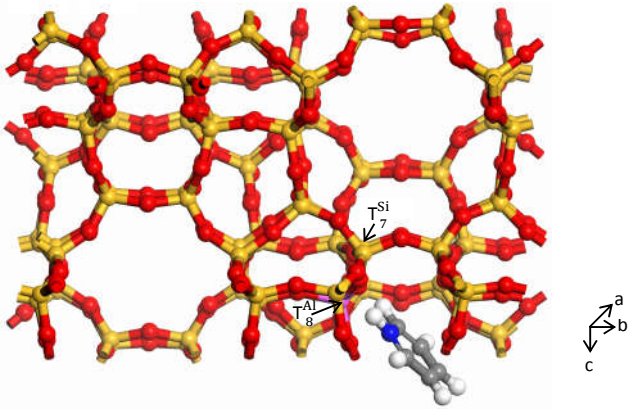


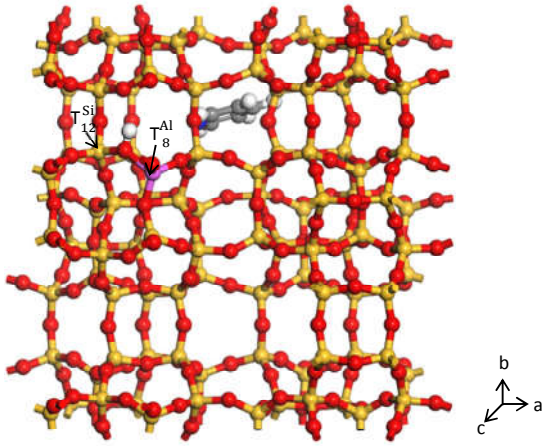
T_8^{Al}-O^H-T_3^{Si}



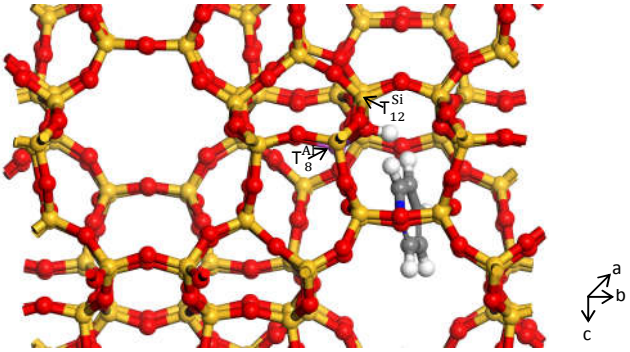


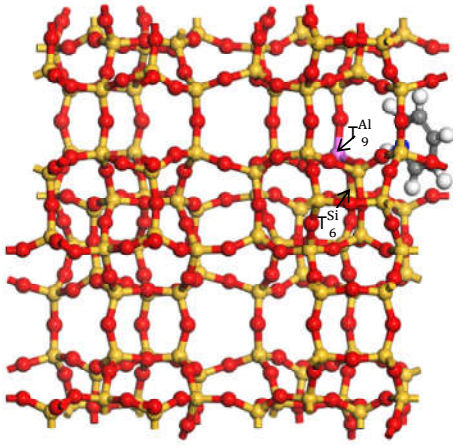
T_8^{Al}-O^H-T_7^{Si}



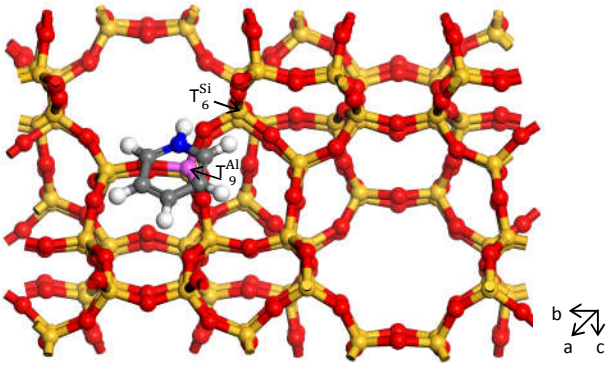


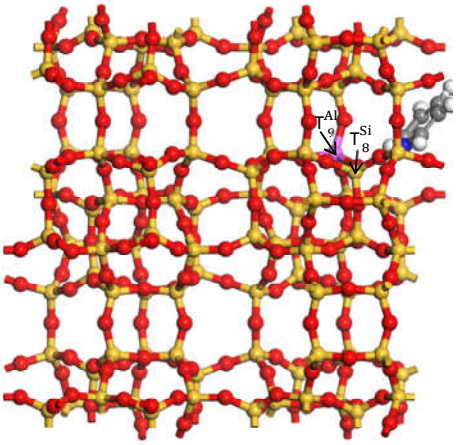
T_8^{Al}-O^H-T_{12}^{Si}



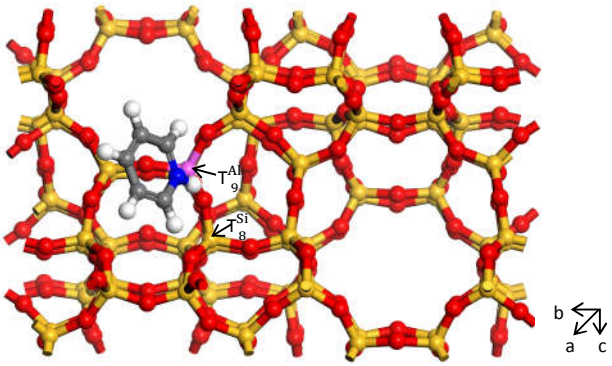


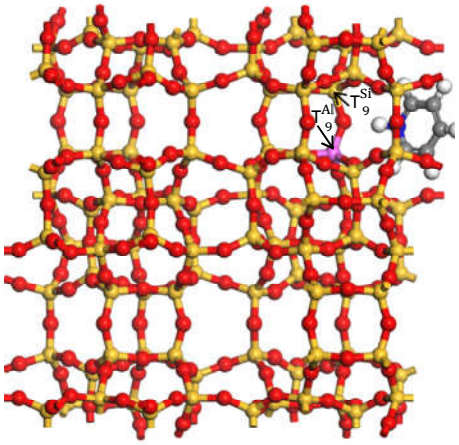
T_9^{Al}-O^H-T_6^{Si}



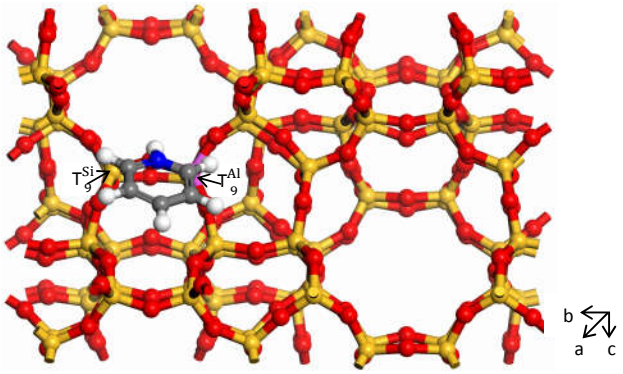


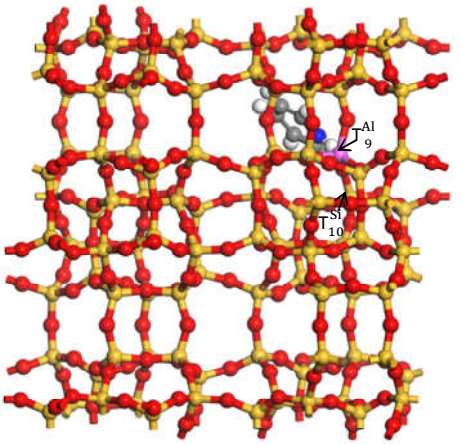
T_9^{Al}-O^H-T_8^{Si}



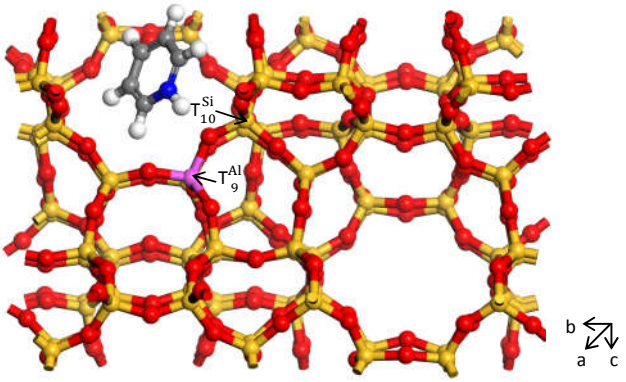


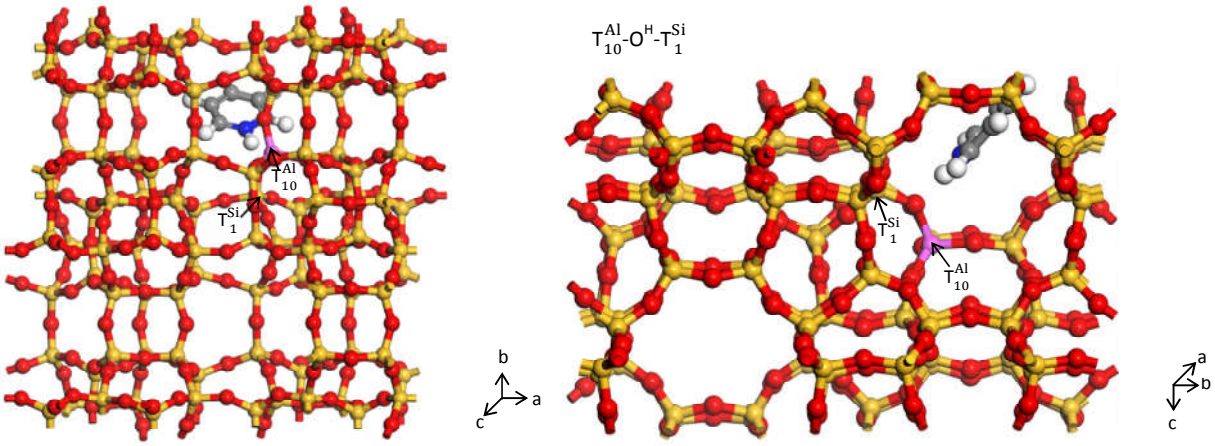
T_9^{Al}-O^H-T_9^{Si}

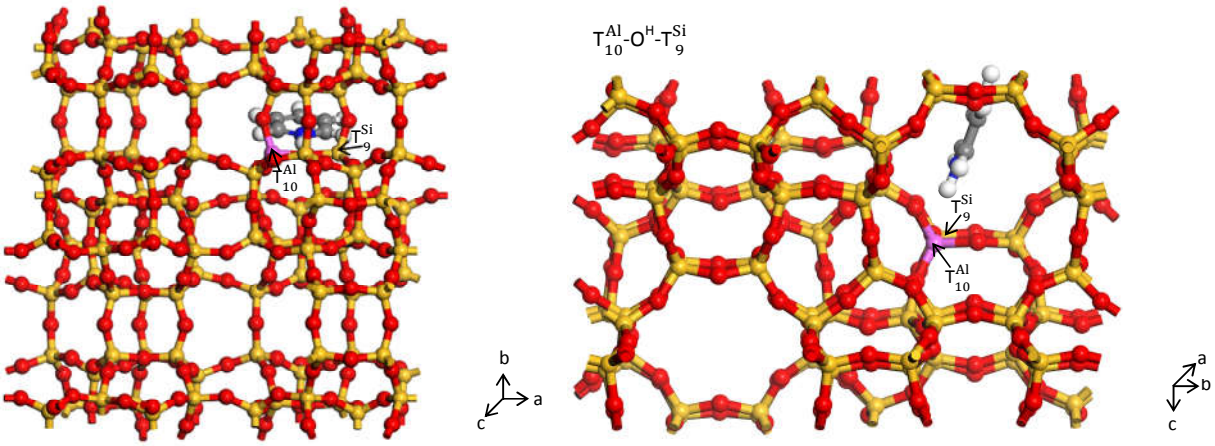


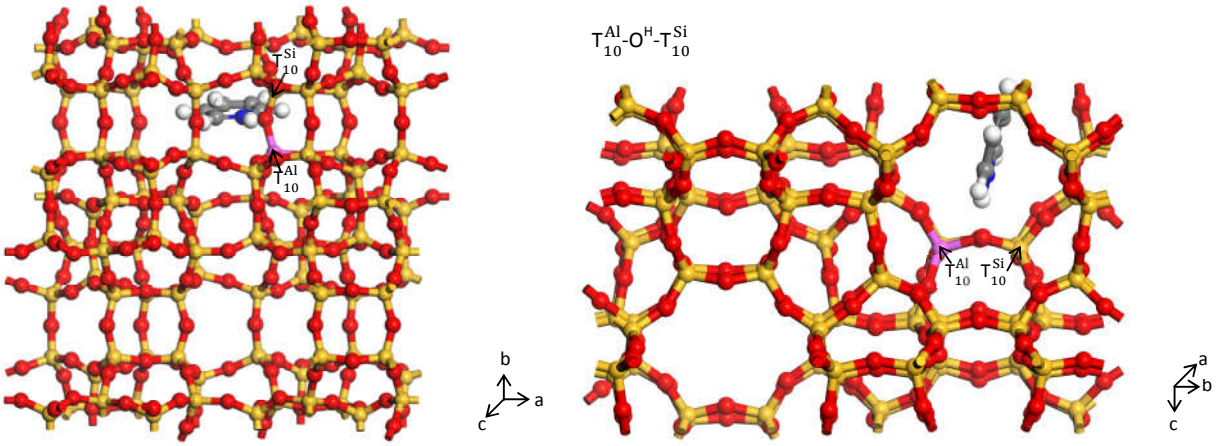


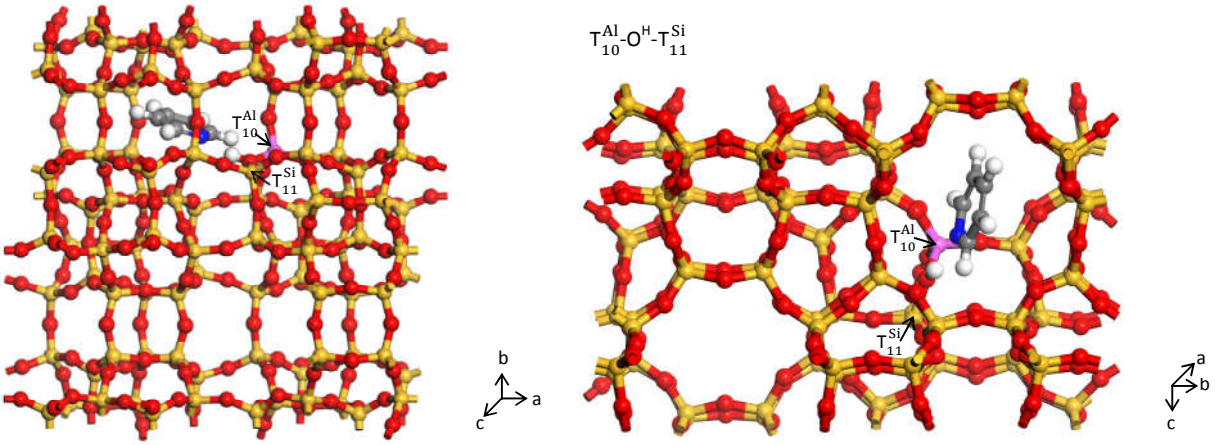
T_9^{Al}-O^H-T_{10}^{Si}

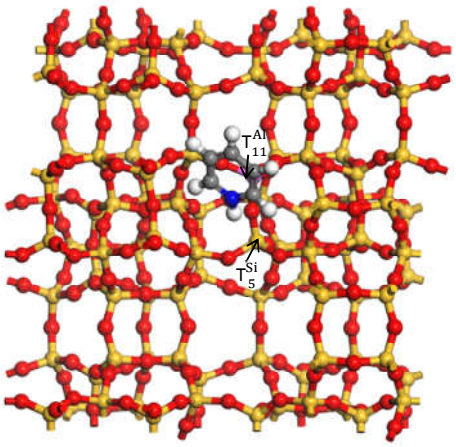



T^{Al}_{10}-O^{H}-T^{Si}_{1}
T^{Al}_{10}
T^{Si}_{1}
b
a
c
a
b
c

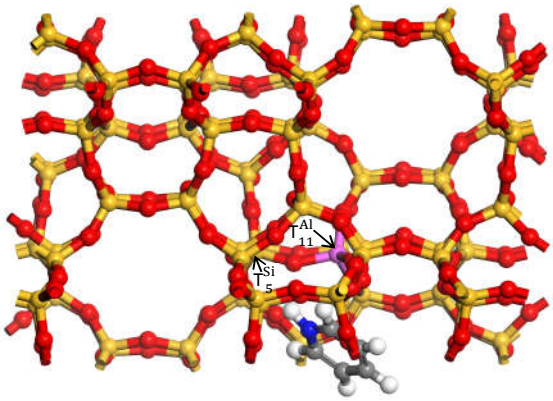

T^{Al}_{10}-O^{H}-T^{Si}_{9}
T^{Si}_{9}
T^{Al}_{10}
b
a
c
a
b
c

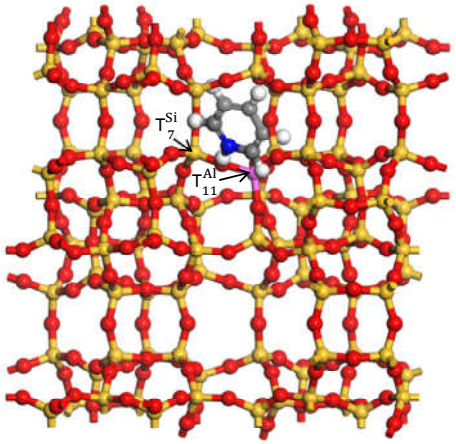

T^{Al}_{10}-O^{H}-T^{Si}_{10}
T^{Si}_{10}
T^{Al}_{10}
T^{Si}_{10}
b
a
c
a
b
c


T^{Al}_{10}-O^{H}-T^{Si}_{11}
T^{Al}_{10}
T^{Si}_{11}
T^{Al}_{10}
T^{Si}_{11}
b
a
c
a
b
c

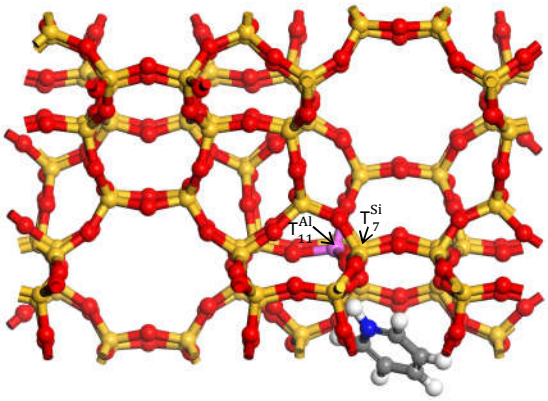


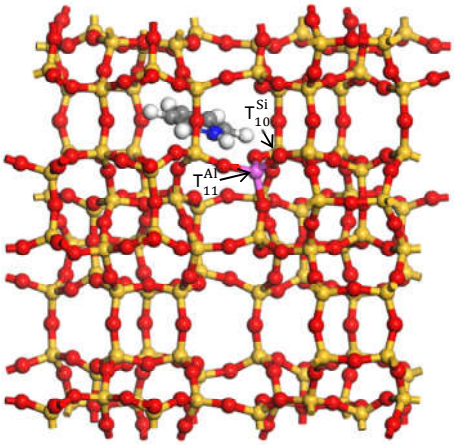
T^{Al}_{11}-O^{H}-T^{Si}_{5}



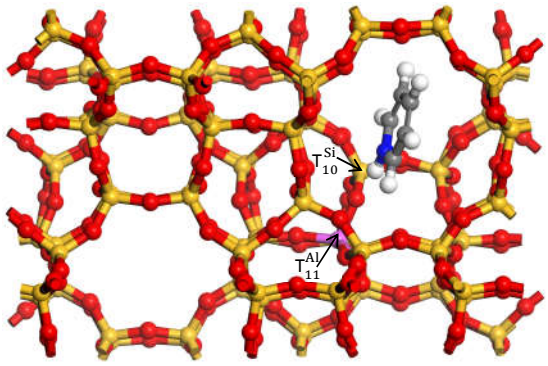


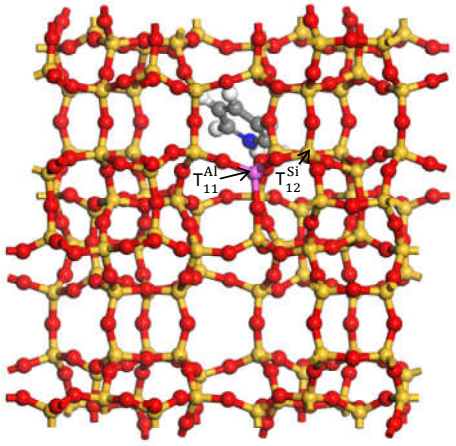
T^{Al}_{11}-O^{H}-T^{Si}_{7}



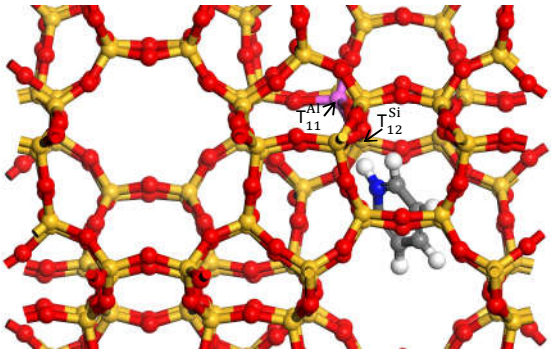


T^{Al}_{11}-O^{H}-T^{Si}_{10}





T^{Al}_{11}-O^{H}-T^{Si}_{12}



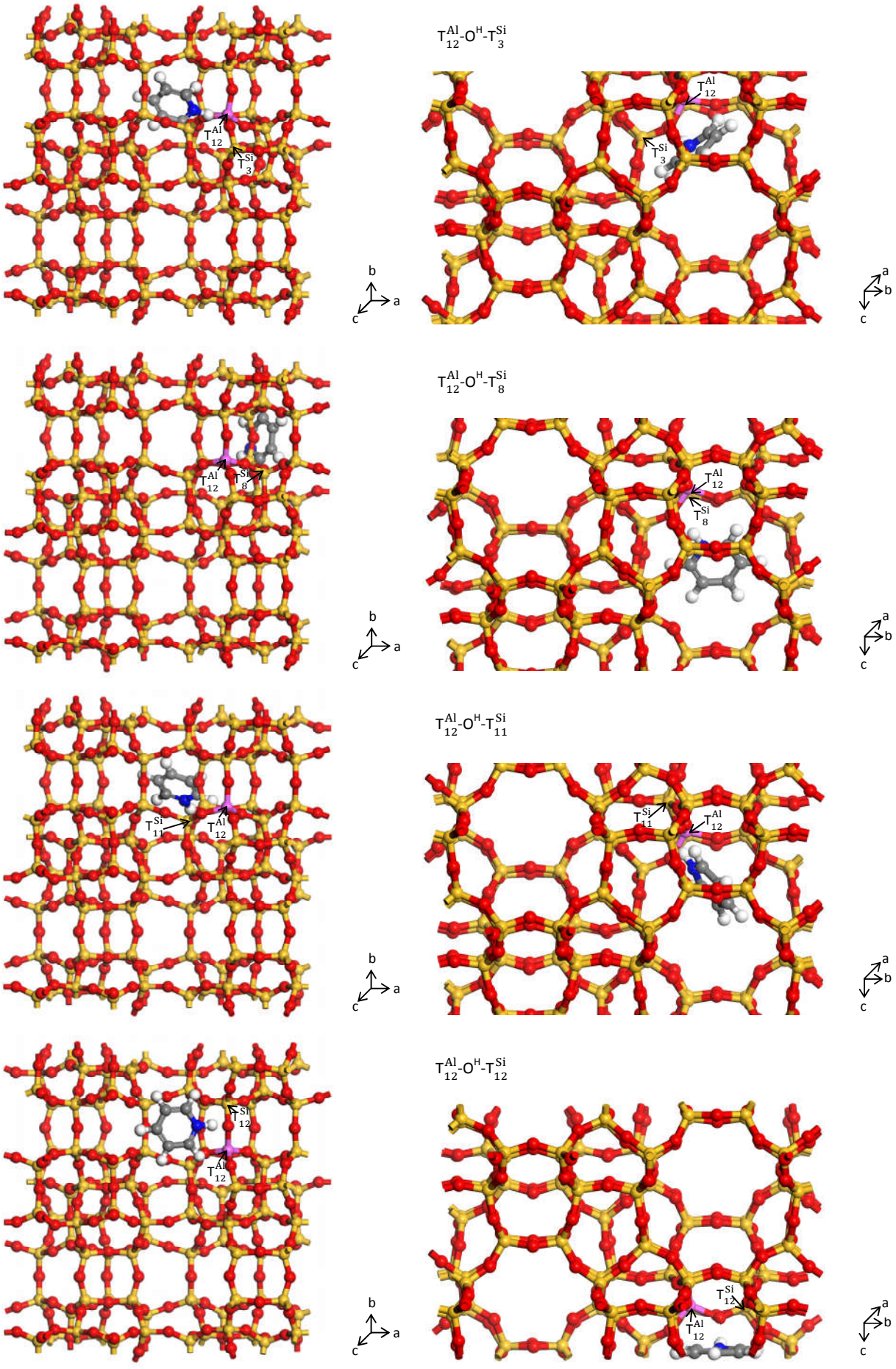
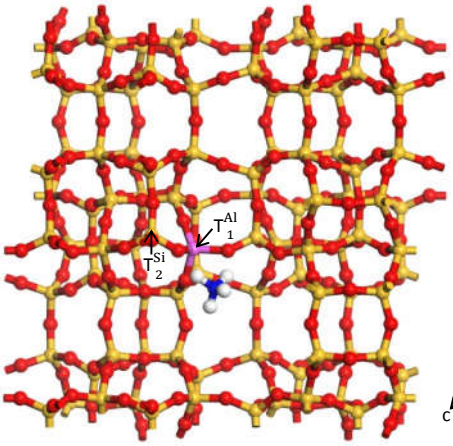
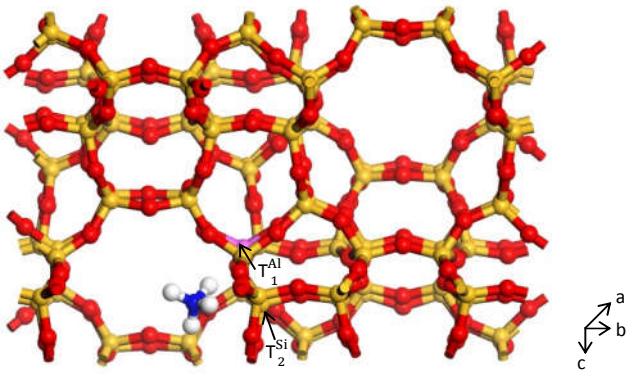
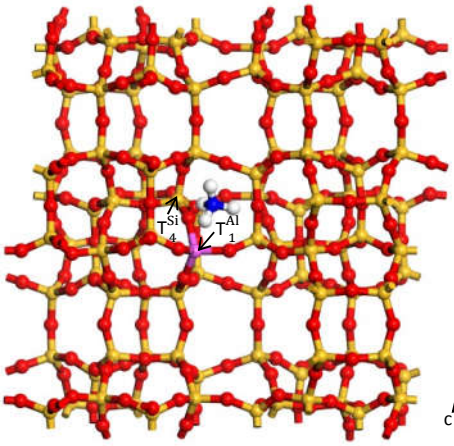


Figure S3 The optimized configurations of pyridine adsorption at all 48 acidic sites of one Al substitution in the framework

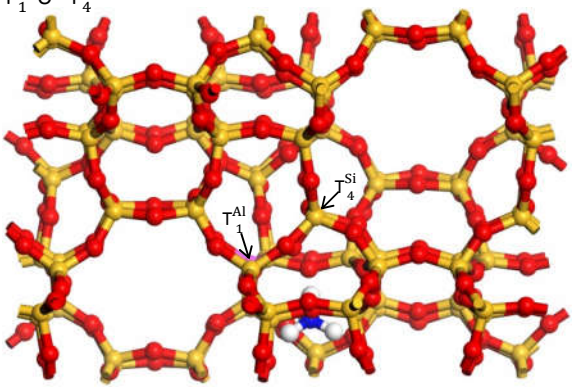


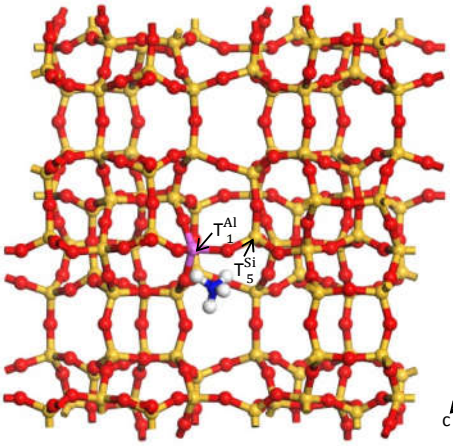
T^{Al}_1-O^H-T^{Si}_2



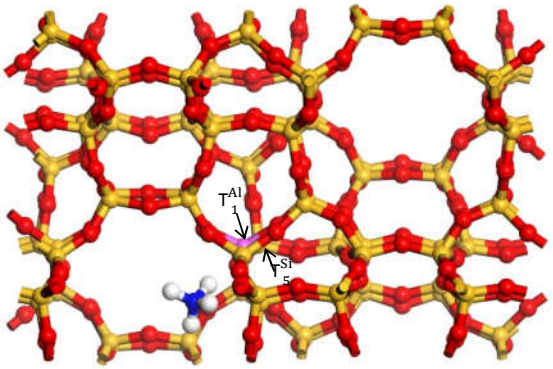


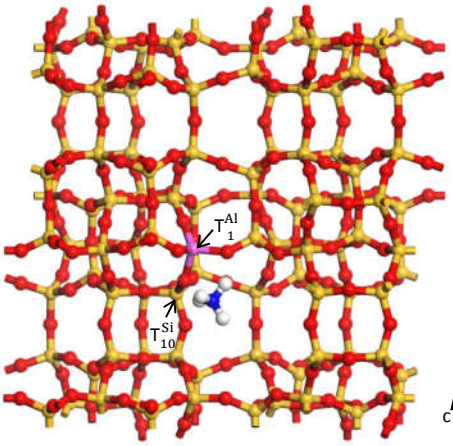
T^{Al}_1-O^H-T^{Si}_4



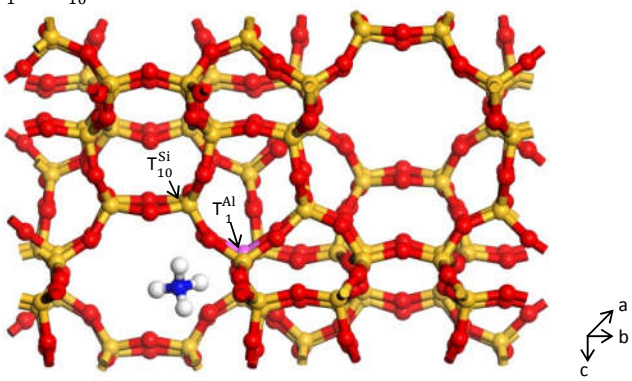


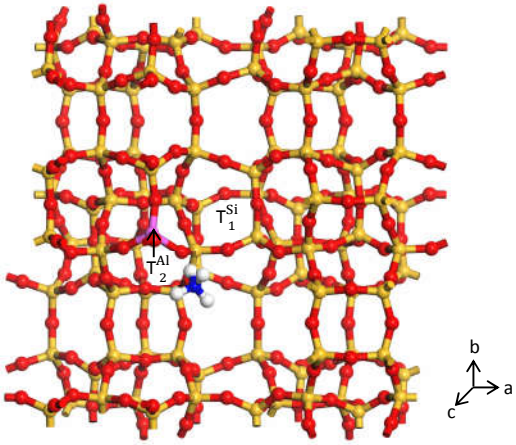
T^{Al}_1-O^H-T^{Si}_5



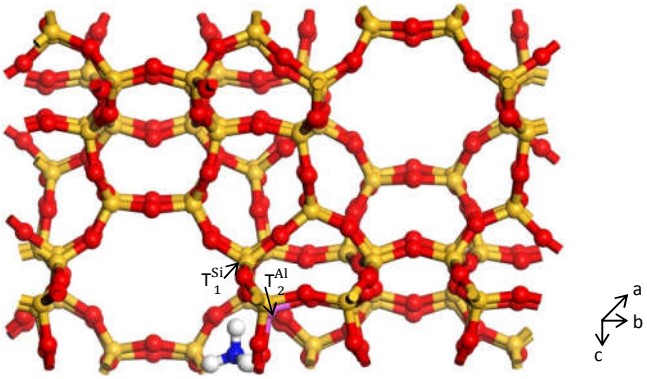


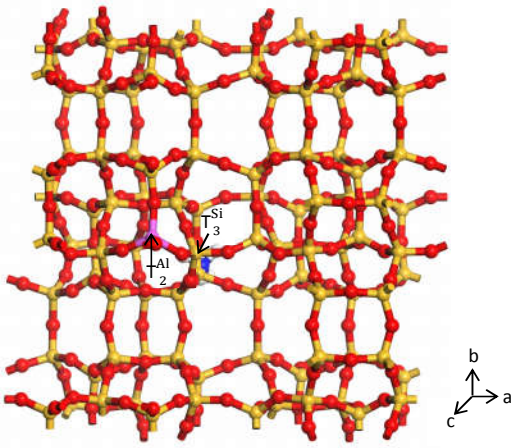
T^{Al}_1-O^H-T^{Si}_{10}



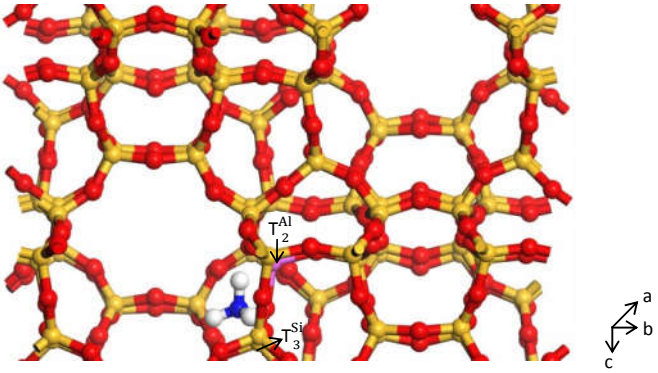


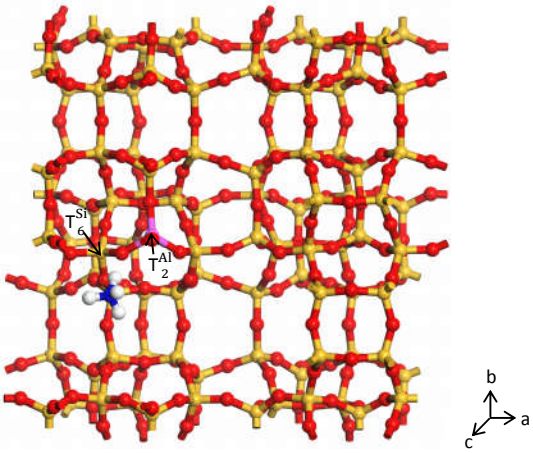
T^{Al}_2-O^H-T^{Si}_1



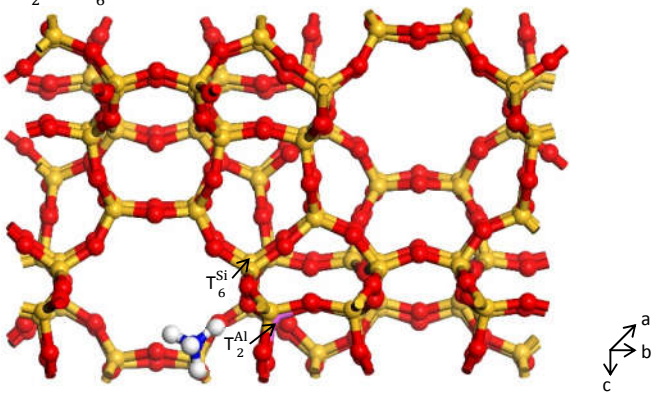


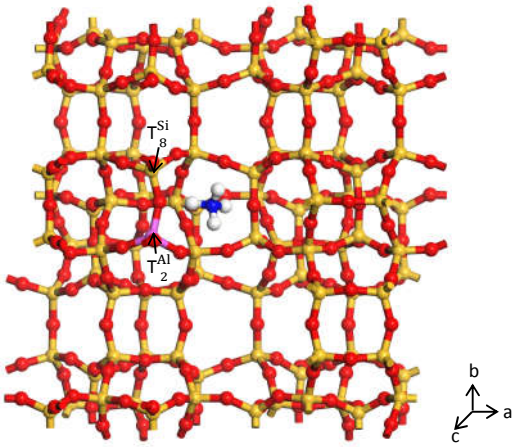
T^{Al}_2-O^H-T^{Si}_3



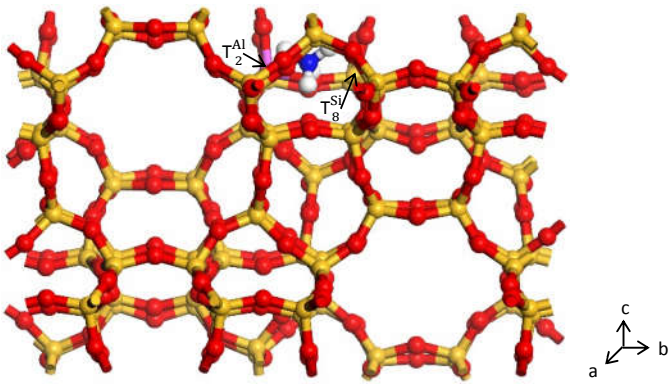


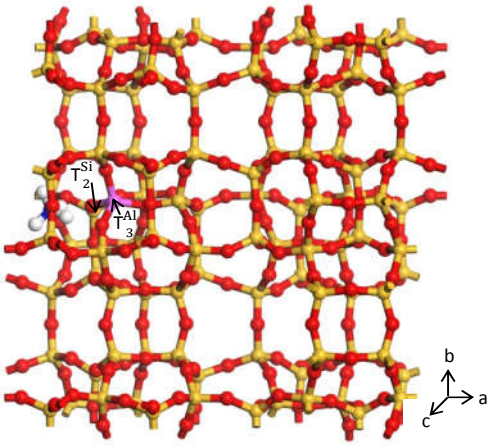
T^{Al}_2-O^H-T^{Si}_6



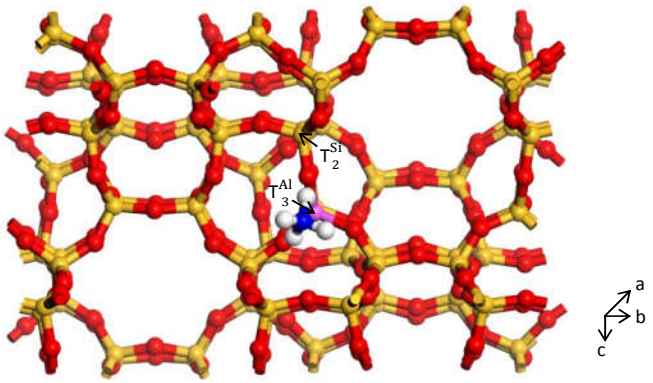


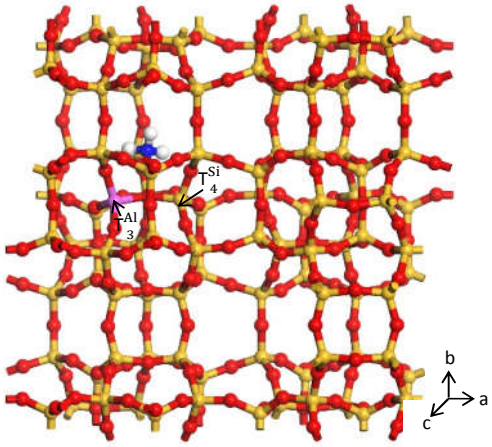
T^{Al}_2-O^H-T^{Si}_8



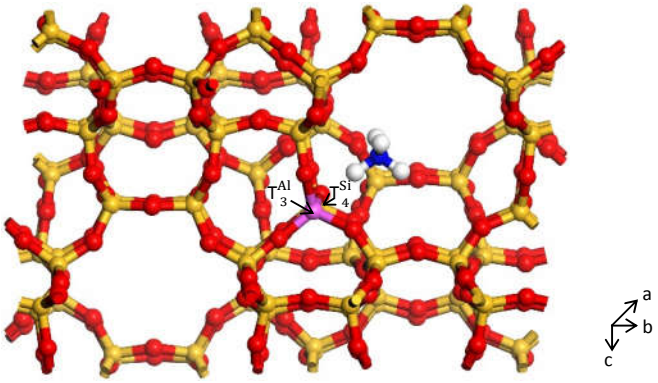


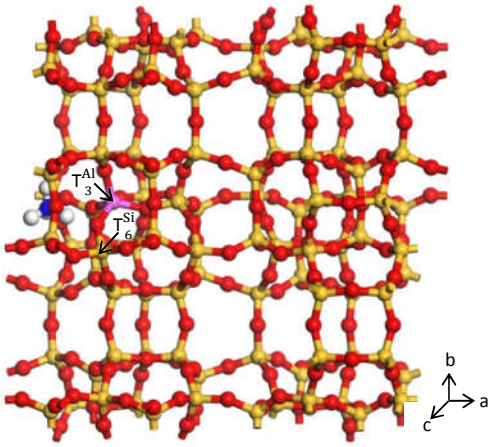
T_3^{Al}-O^H-T_2^{Si}



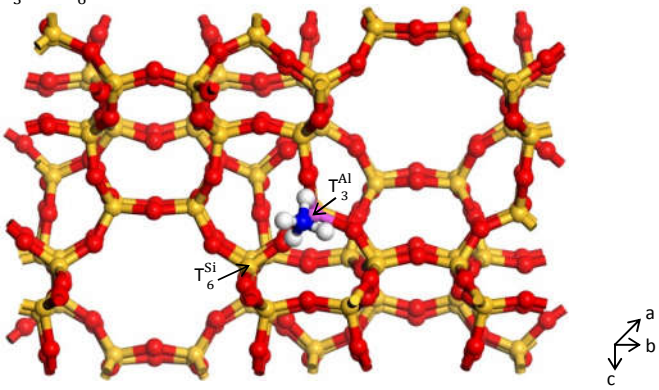


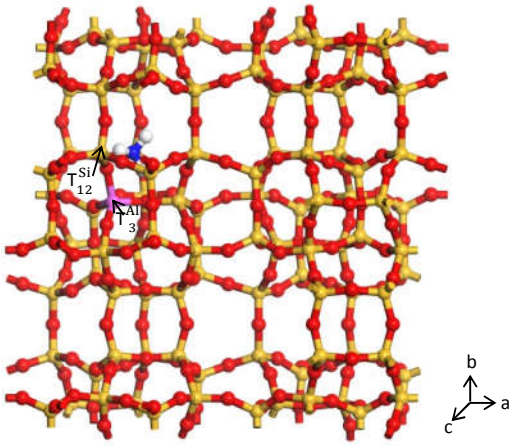
T_3^{Al}-O^H-T_4^{Si}



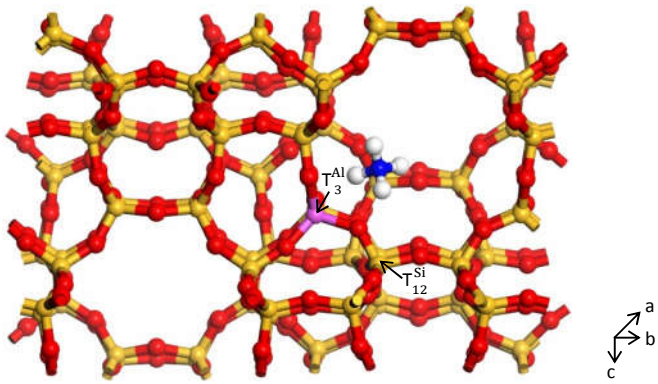


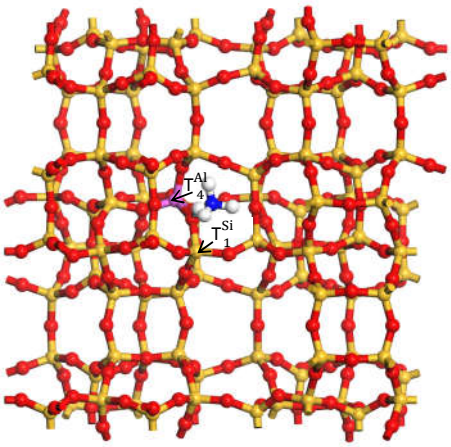
T_3^{Al}-O^H-T_6^{Si}



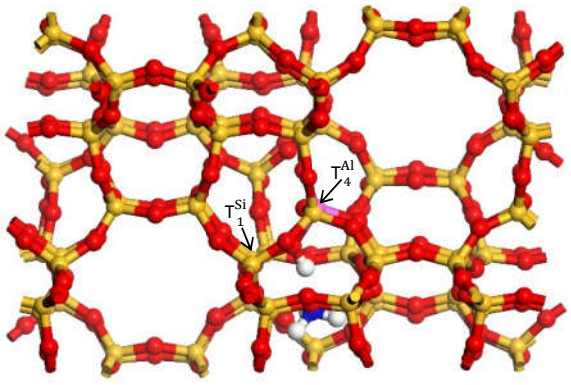


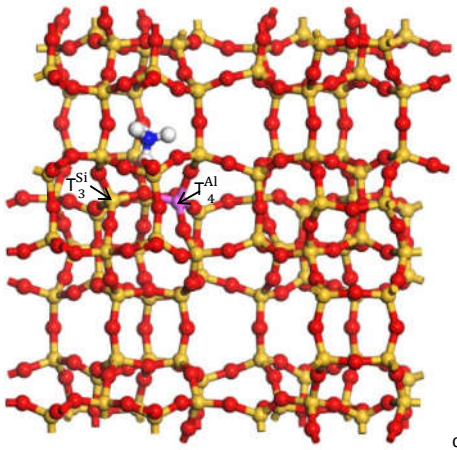
T_3^{Al}-O^H-T_{12}^{Si}



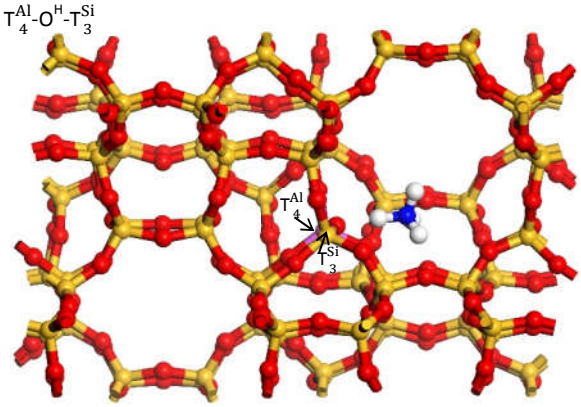


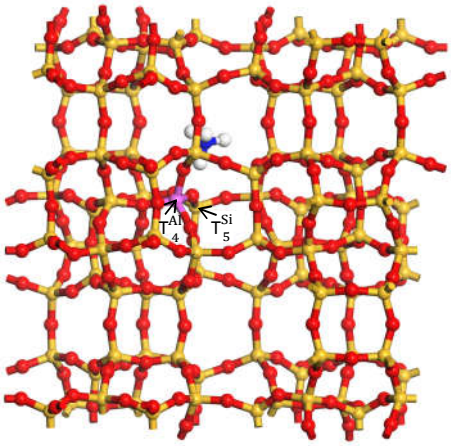
T^{Al}_{4}-O^{H}-T^{Si}_{1}



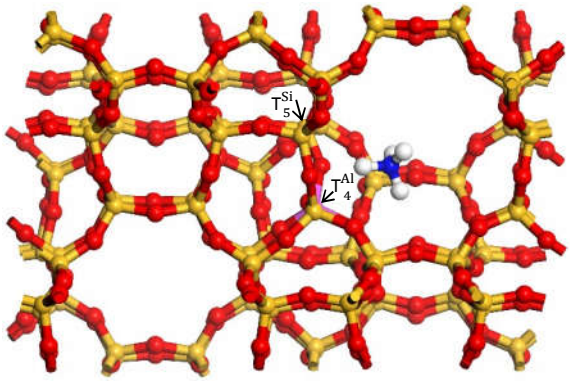


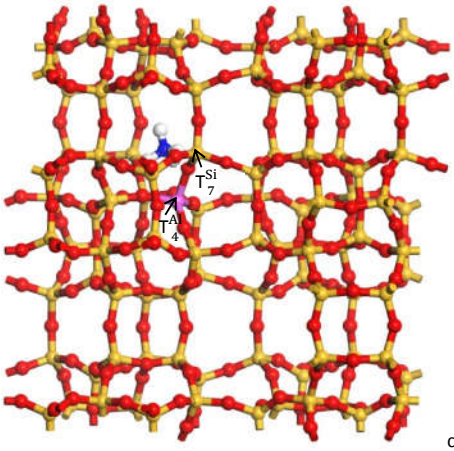
T^{Al}_{4}-O^{H}-T^{Si}_{3}



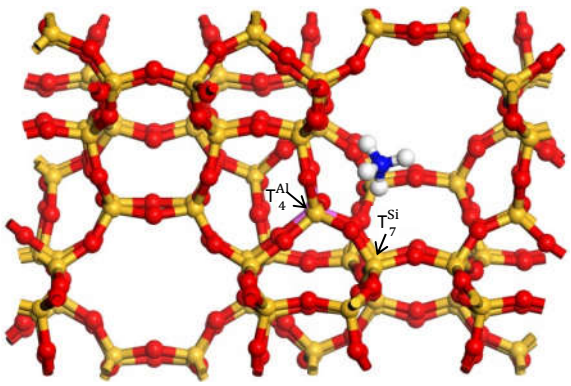


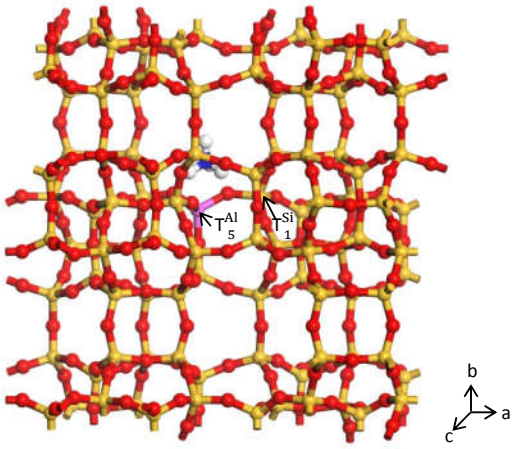
T^{Al}_{4}-O^{H}-T^{Si}_{5}



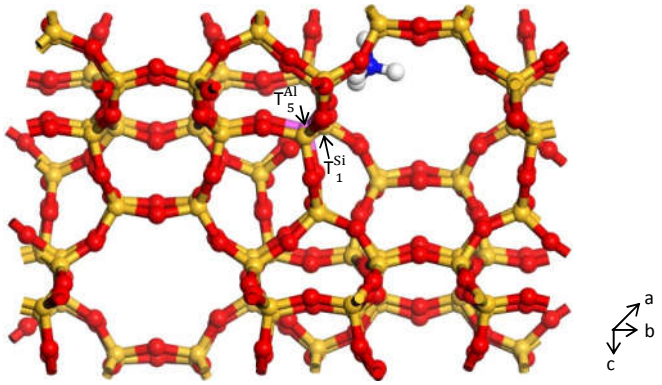


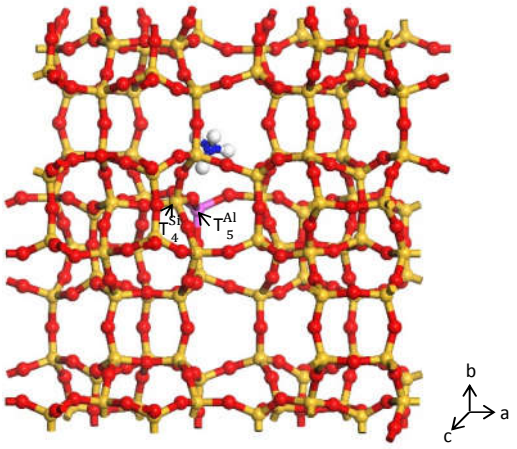
T^{Al}_{4}-O^{H}-T^{Si}_{7}



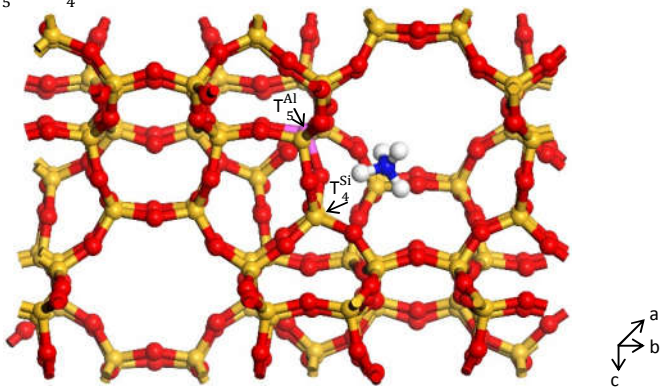


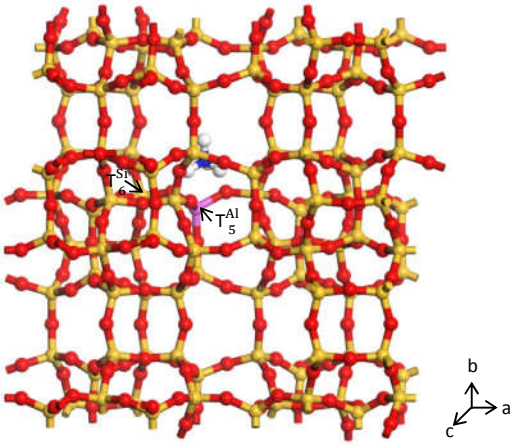
T^{Al}_5-O^H-T^{Si}_1



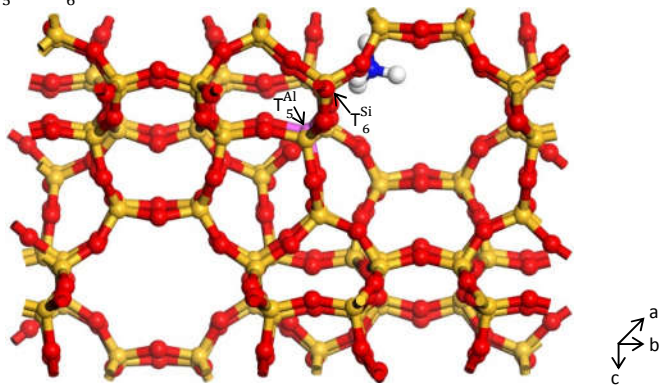


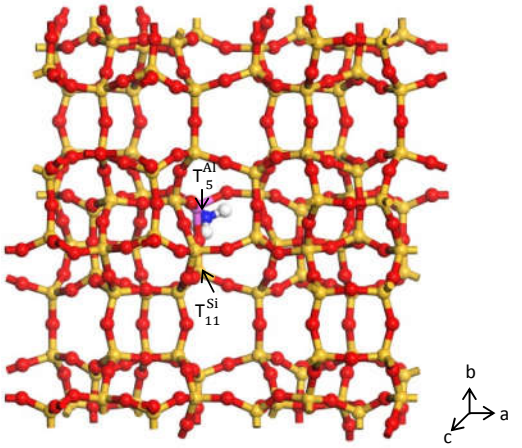
T^{Al}_5-O^H-T^{Si}_4



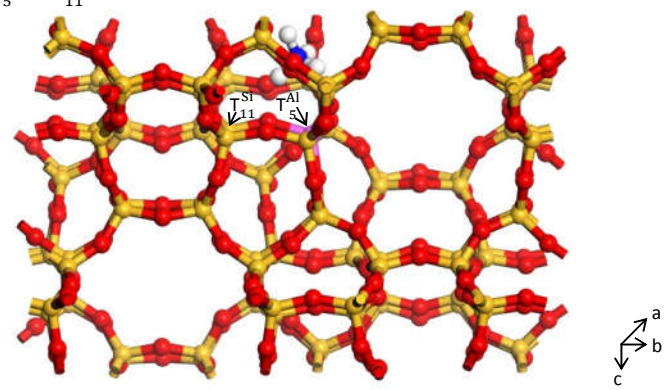


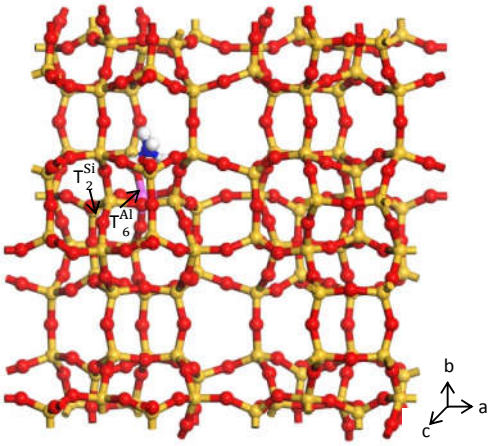
T^{Al}_5-O^H-T^{Si}_6



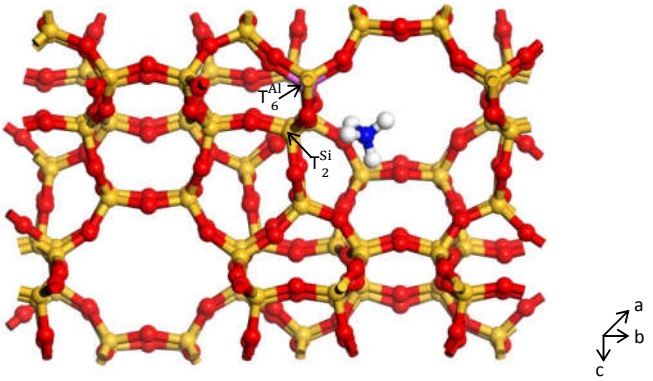


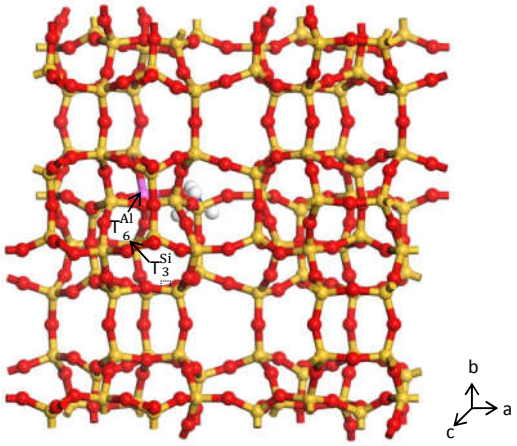
T^{Al}_5-O^H-T^{Si}_{11}



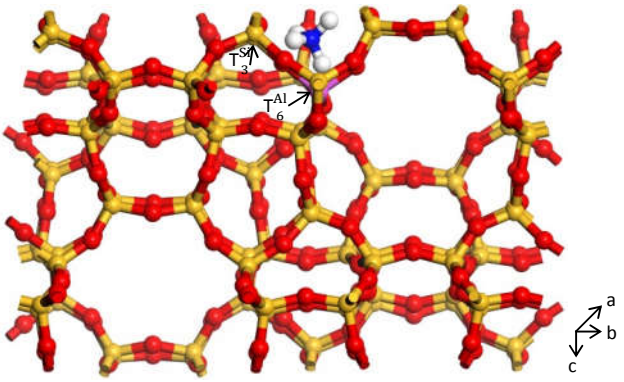


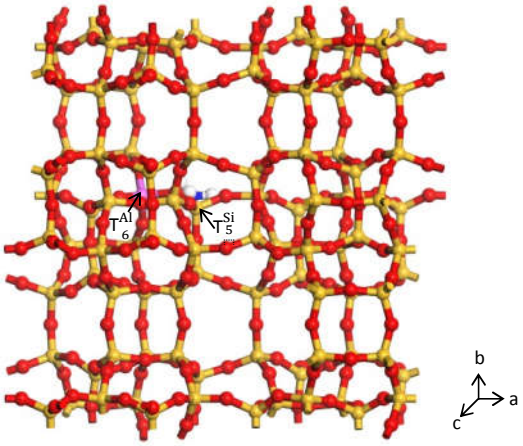
T_6^{Al}-O^H-T_2^{Si}



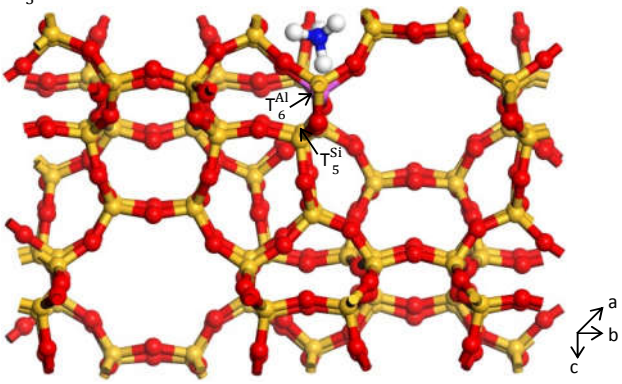


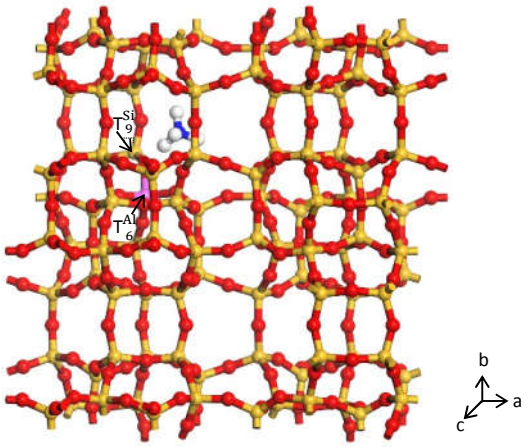
T_6^{Al}-O^H-T_3^{Si}



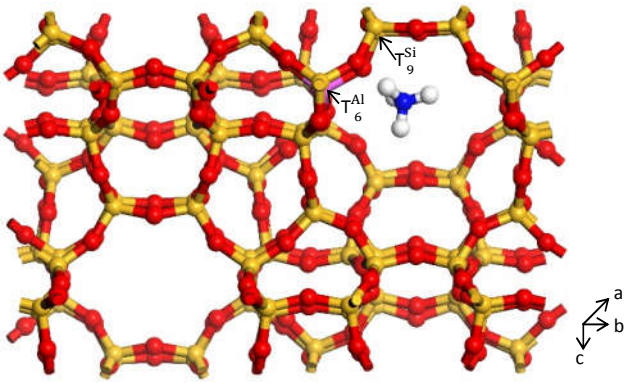


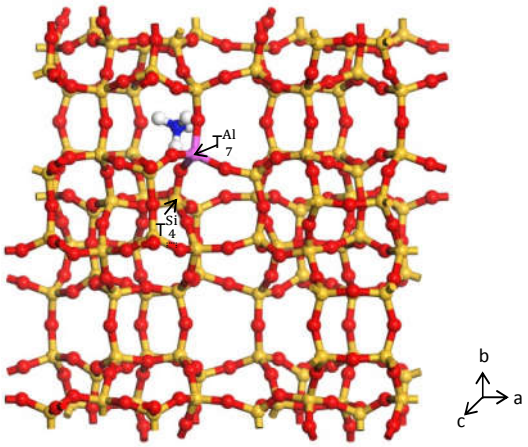
T_6^{Al}-O^H-T_5^{Si}



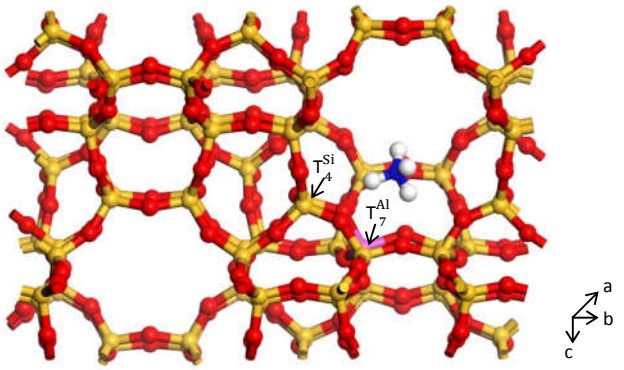


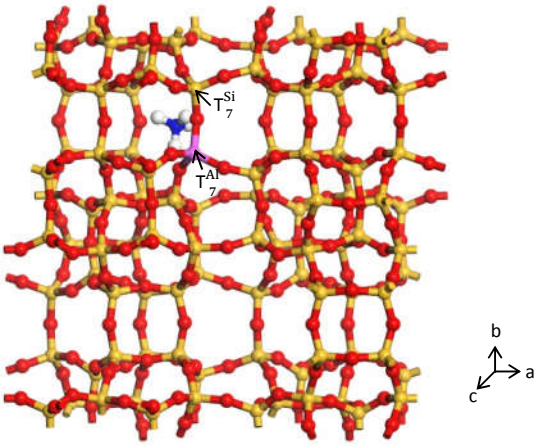
T_6^{Al}-O^H-T_9^{Si}



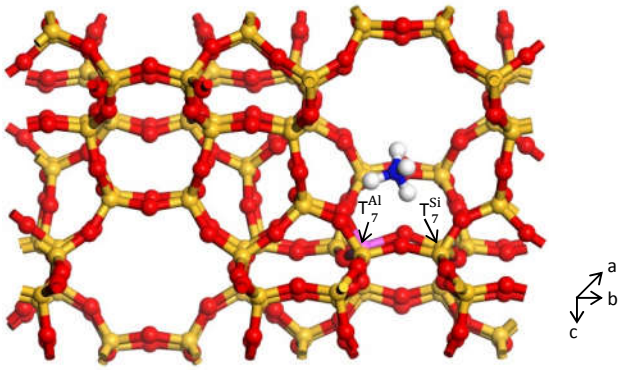


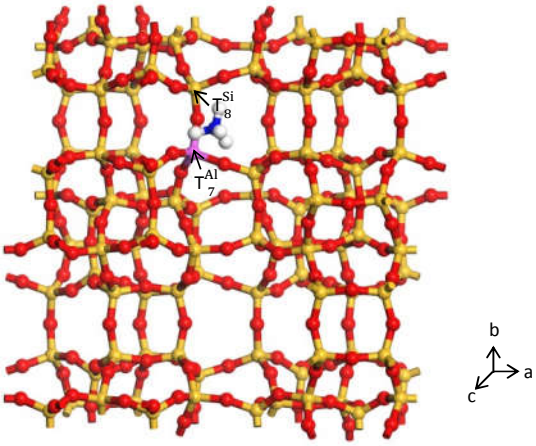
T^{Al}_7-O^H-T^{Si}_4



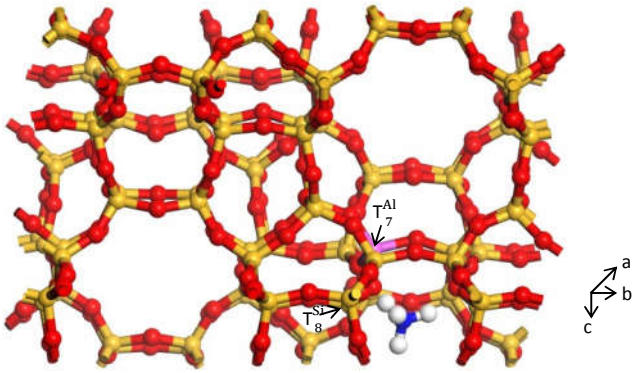


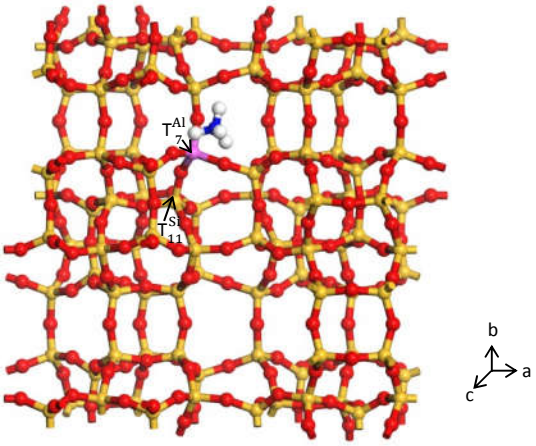
T^{Al}_7-O^H-T^{Si}_7



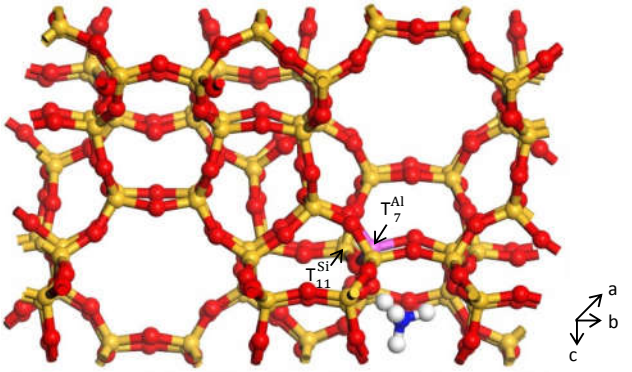


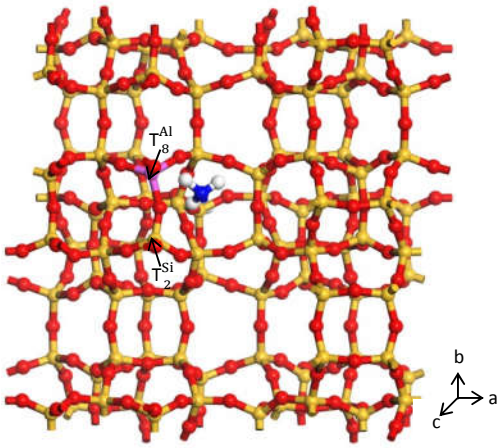
T^{Al}_7-O^H-T^{Si}_8



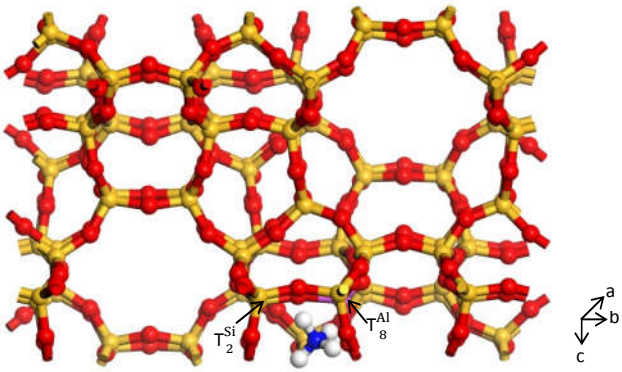


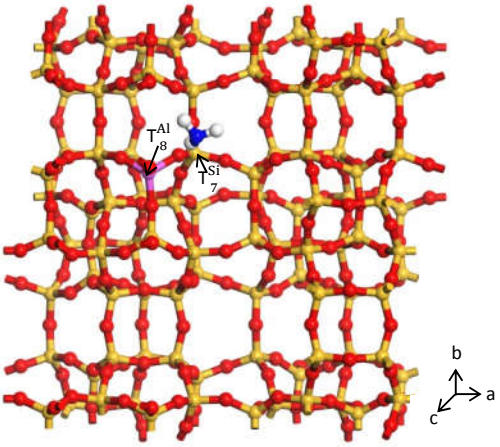
T^{Al}_7-O^H-T^{Si}_{11}



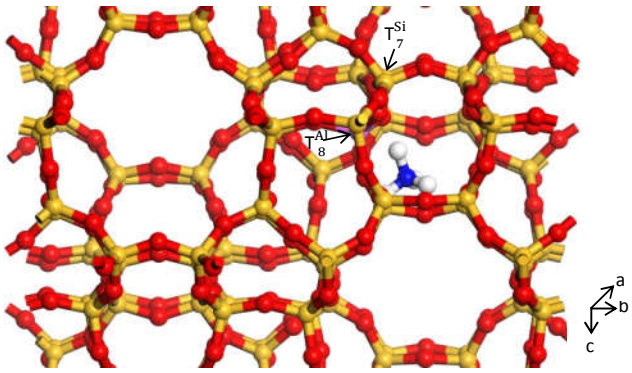


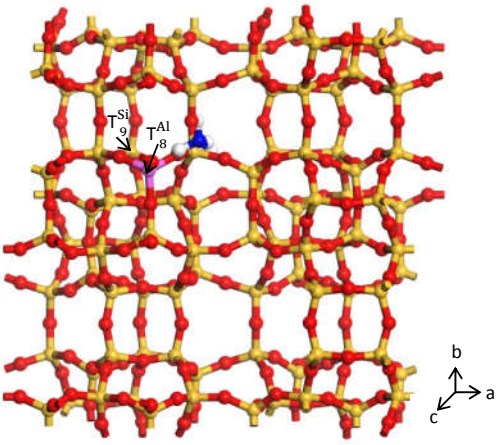
T^{Al}_{8}-O^{H}-T^{Si}_{2}



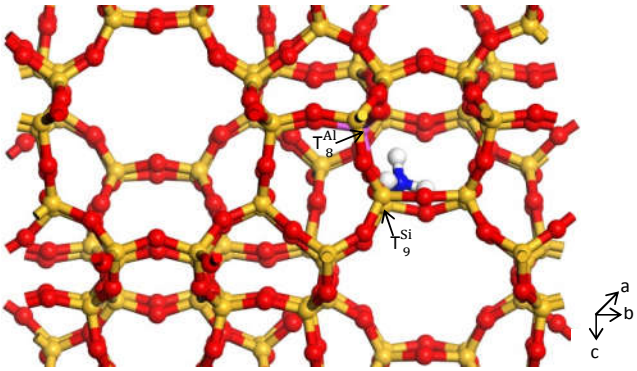


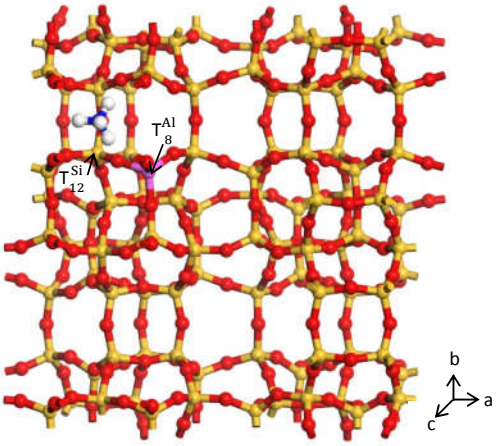
T^{Al}_{8}-O^{H}-T^{Si}_{7}



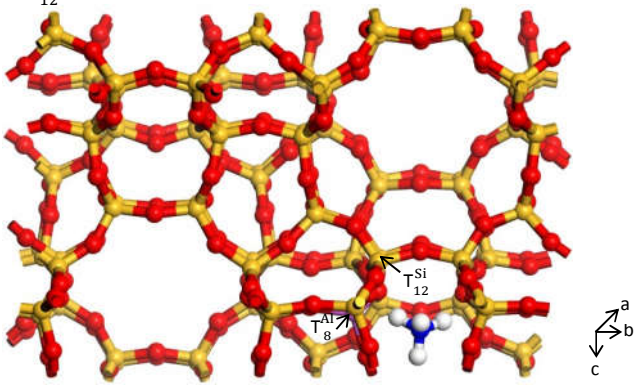


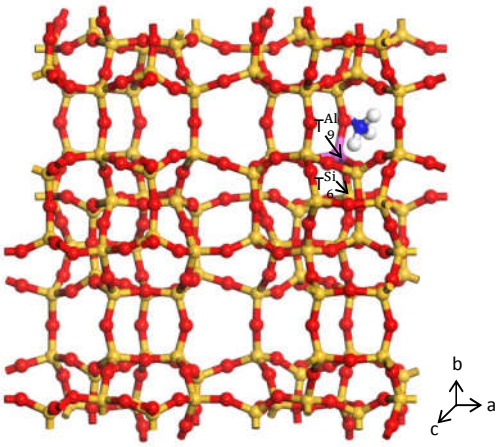
T^{Al}_{8}-O^{H}-T^{Si}_{9}



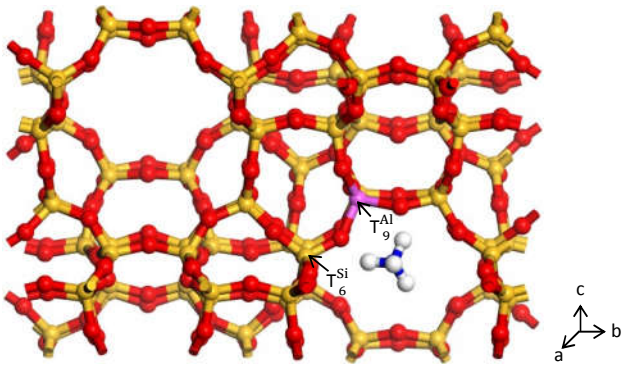


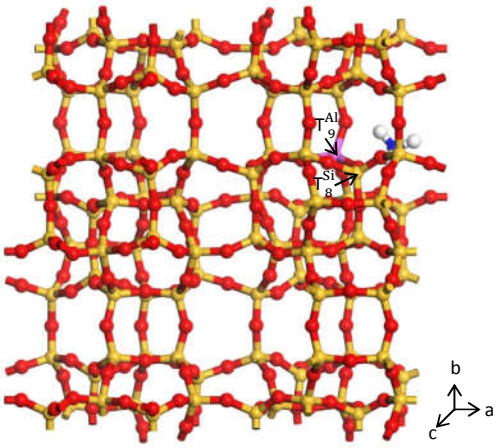
T^{Al}_{8}-O^{H}-T^{Si}_{12}



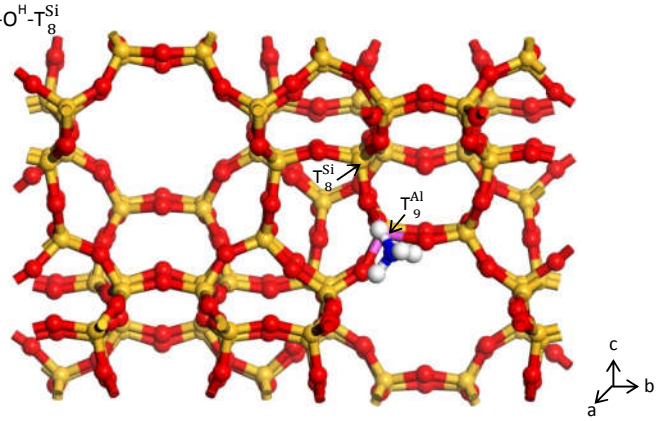


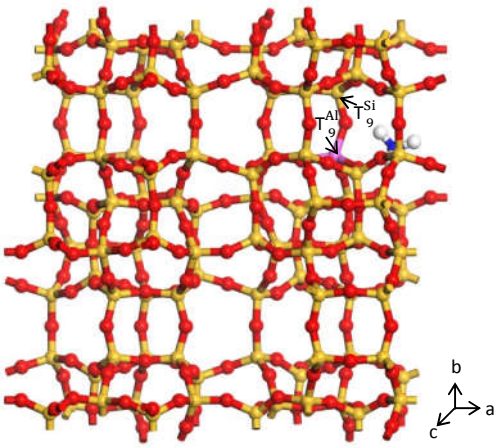
T^{Al}_{9}-O^{H}-T^{Si}_{6}



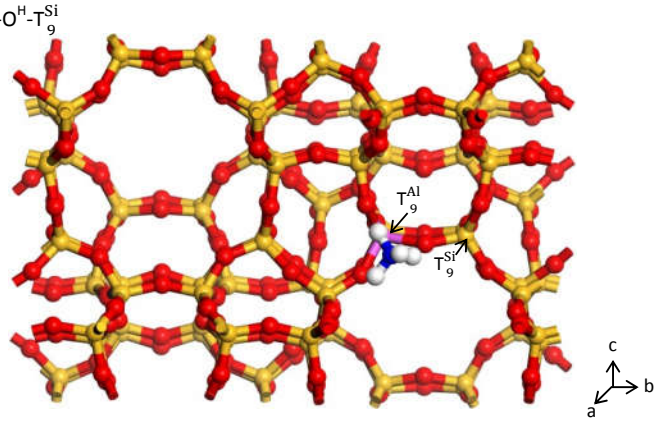


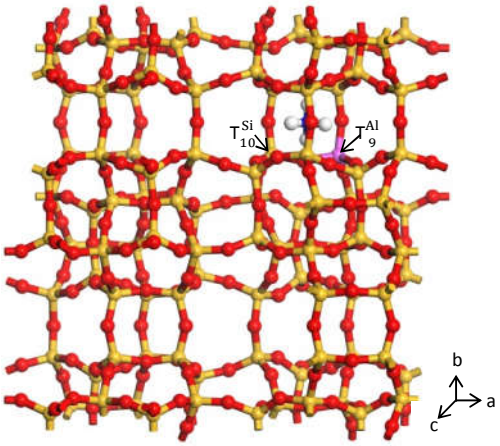
T^{Al}_{9}-O^{H}-T^{Si}_{8}



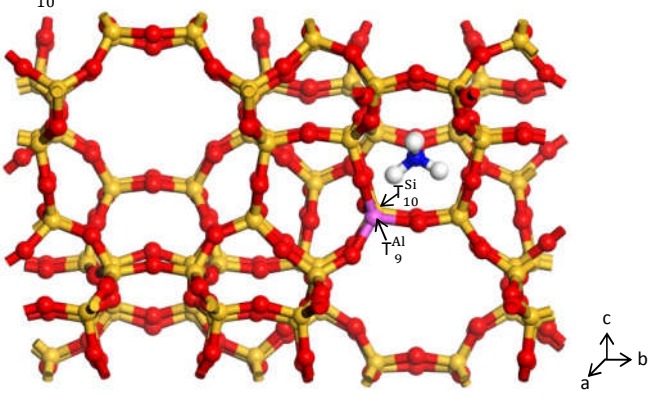


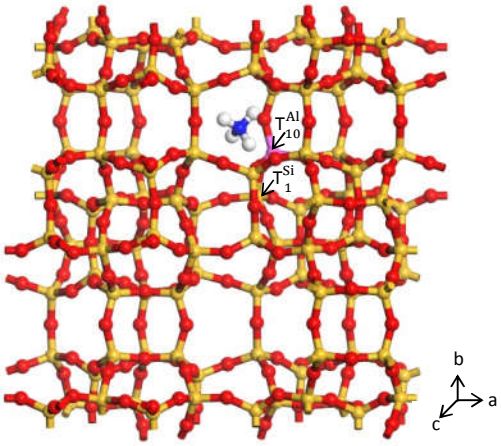
T^{Al}_{9}-O^{H}-T^{Si}_{9}



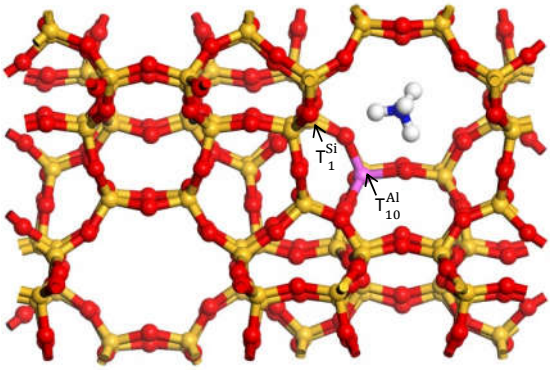


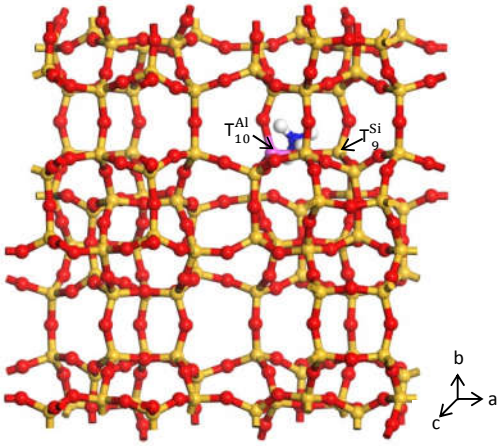
T^{Al}_{9}-O^{H}-T^{Si}_{10}



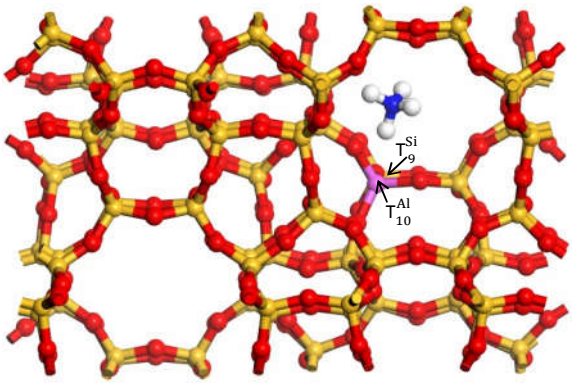


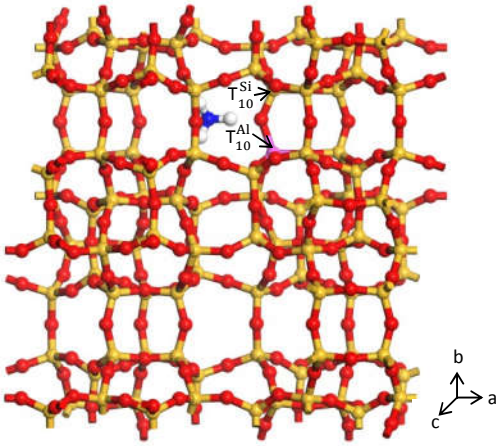
T^{Al}_{10}-O^{H}-T^{Si}_{1}



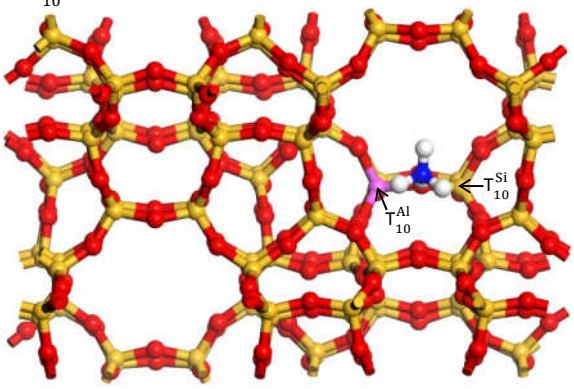


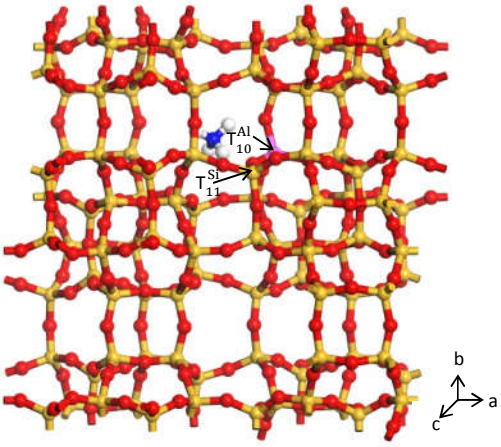
T^{Al}_{10}-O^{H}-T^{Si}_{9}



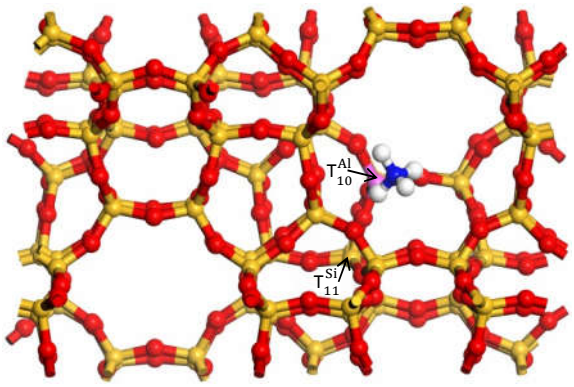


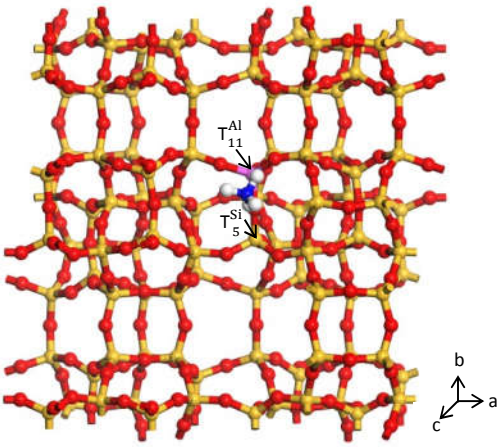
T^{Al}_{10}-O^{H}-T^{Si}_{10}



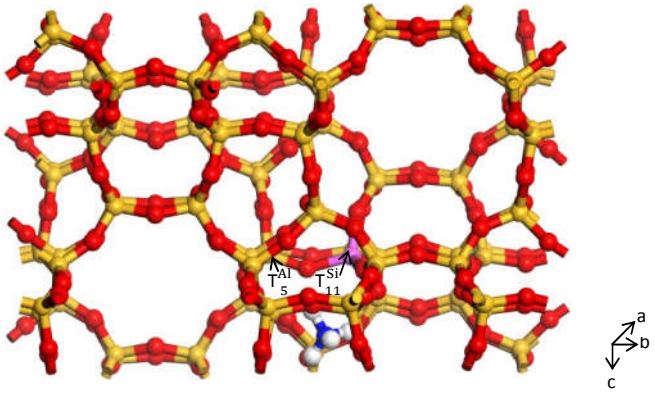


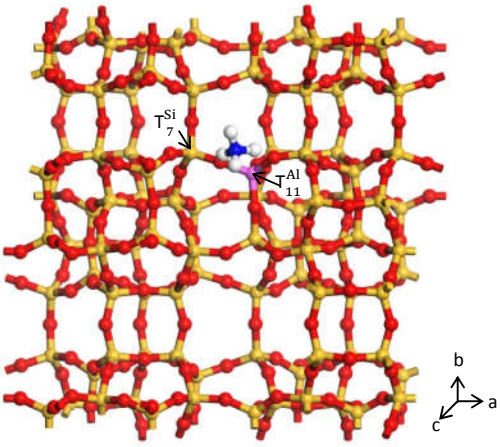
T^{Al}_{10}-O^{H}-T^{Si}_{11}



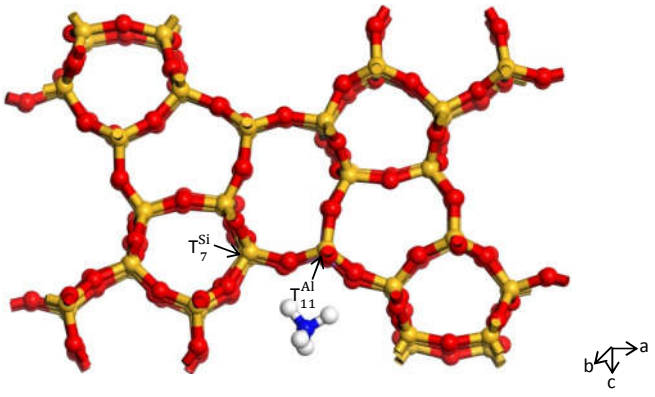


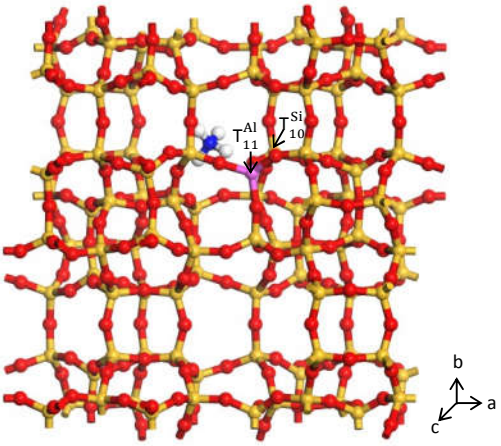
T_{11}^{Al}-O^{H}-T_{5}^{Si}



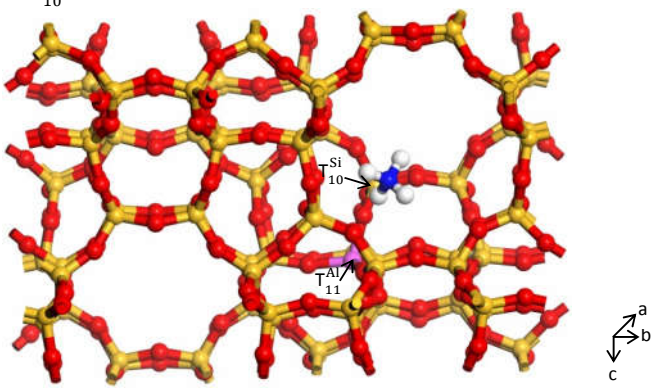


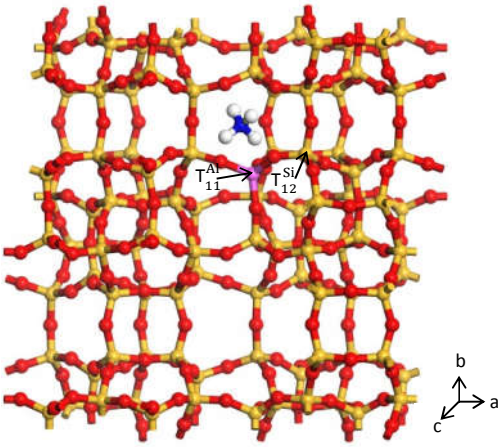
T_{11}^{Al}-O^{H}-T_{7}^{Si}



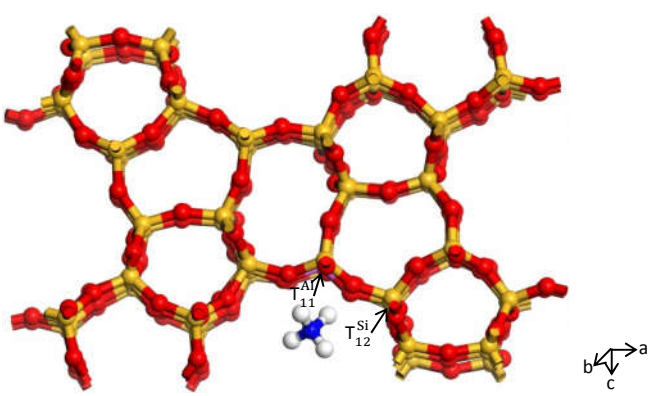


T_{11}^{Al}-O^{H}-T_{10}^{Si}





T_{11}^{Al}-O^{H}-T_{12}^{Si}



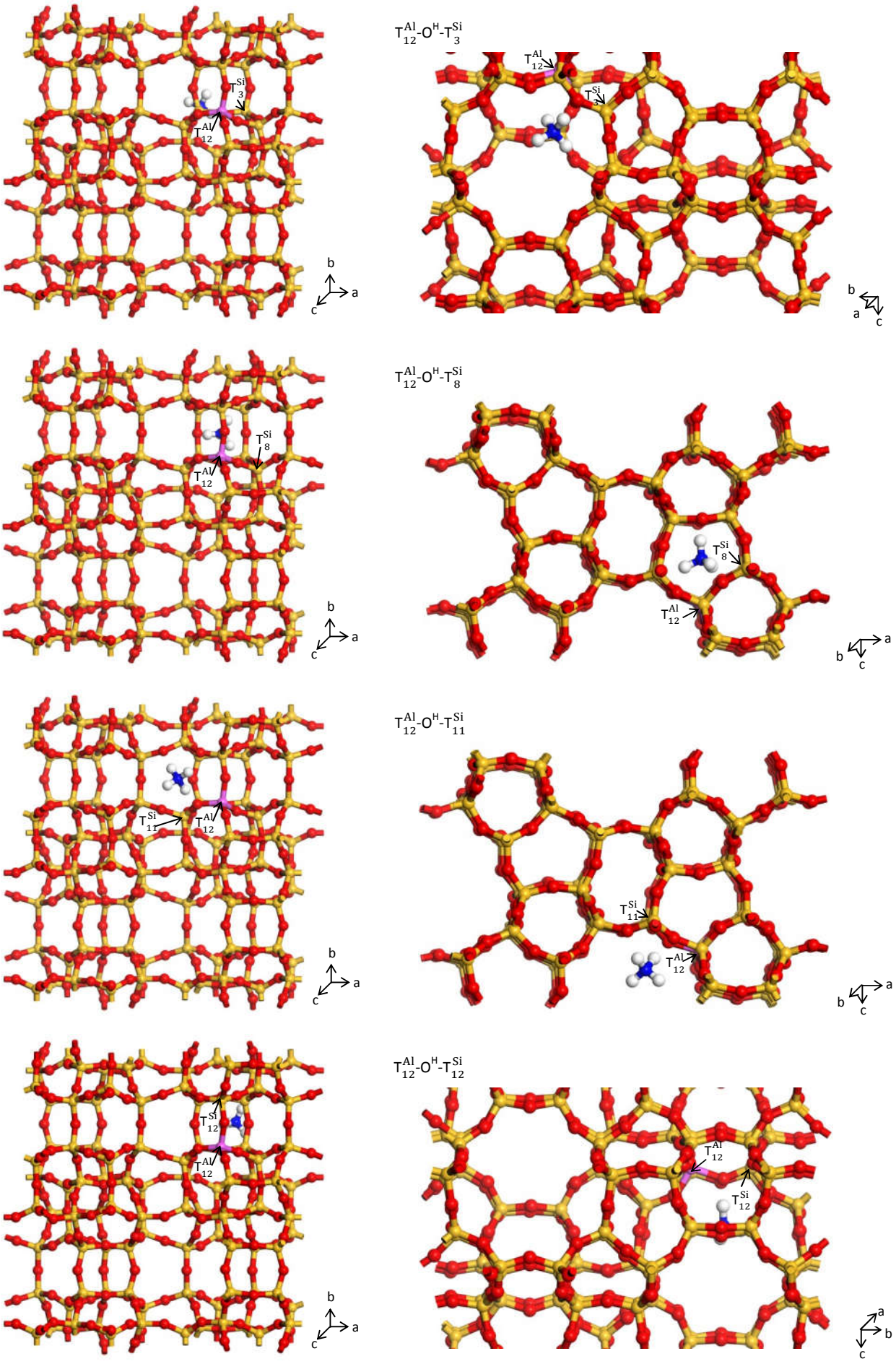
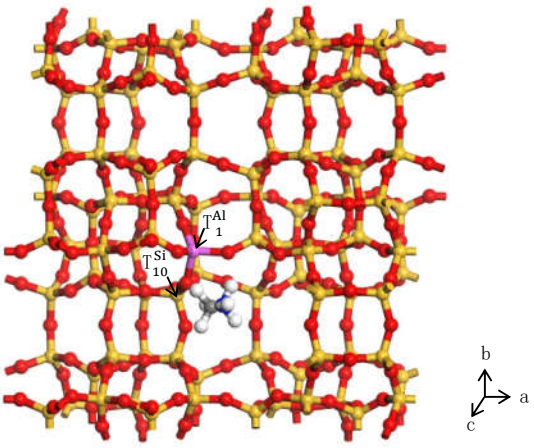
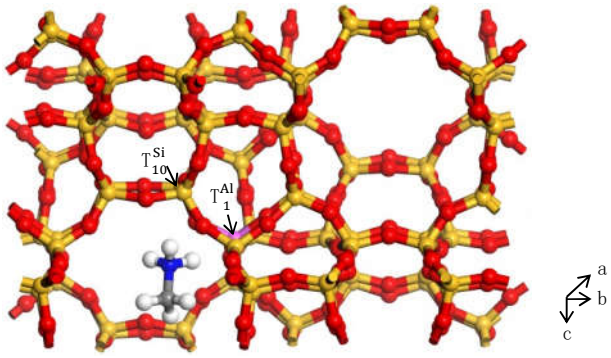
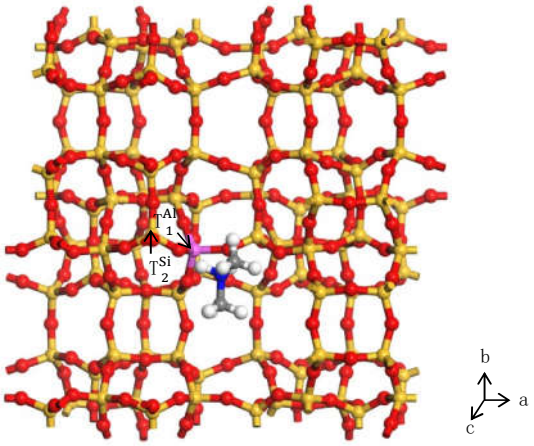


Figure S4 The optimized configurations of ammonia adsorption at all 48 acidic sites of one Al substitution in the framework

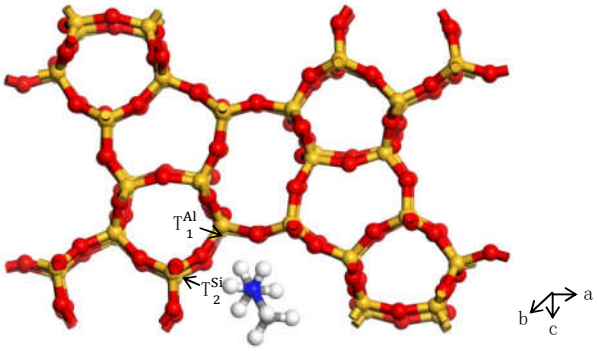


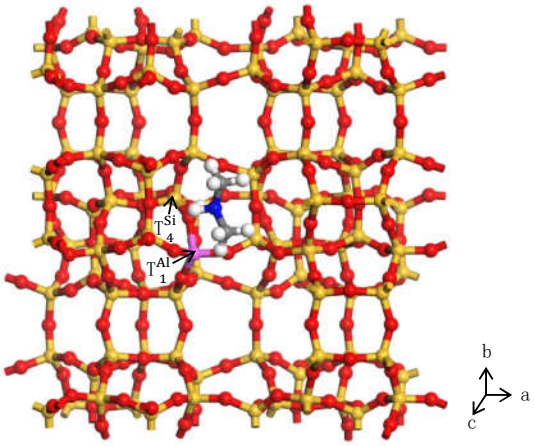
T^{Al}_{1}–O^{II}–T^{Si}_{10}



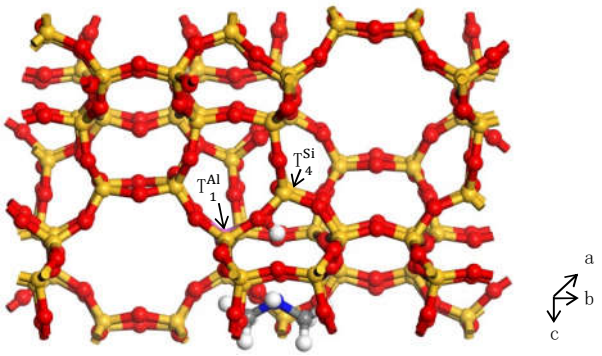


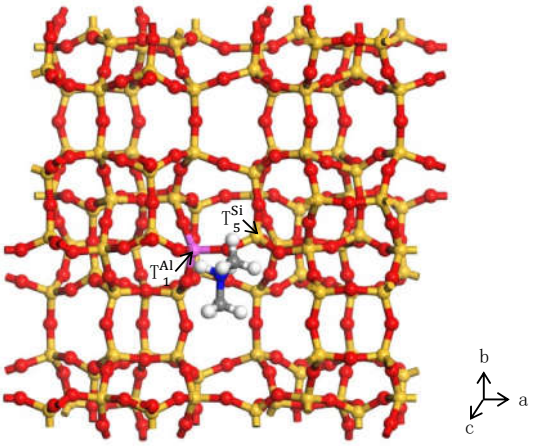
T^{Al}_{1}–O^{II}–T^{Si}_{2}



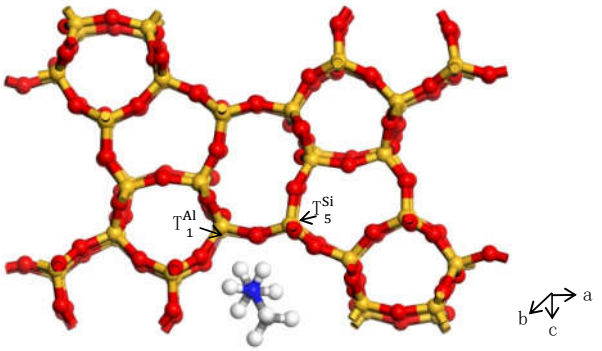


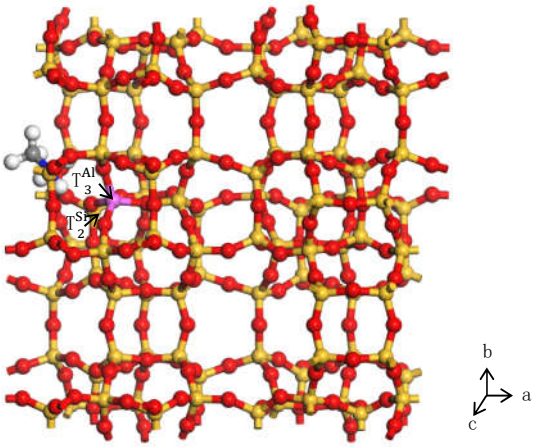
T^{Al}_{1}–O^{II}–T^{Si}_{4}



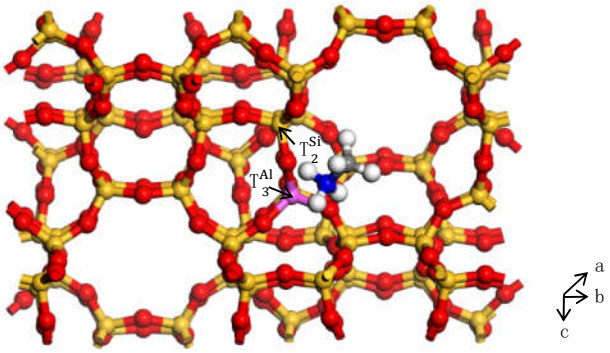


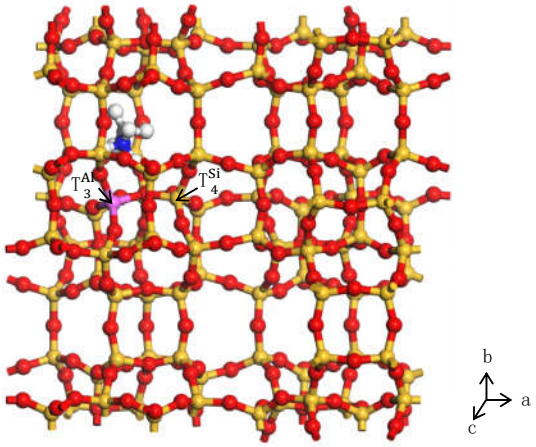
T^{Al}_{1}–O^{II}–T^{Si}_{5}



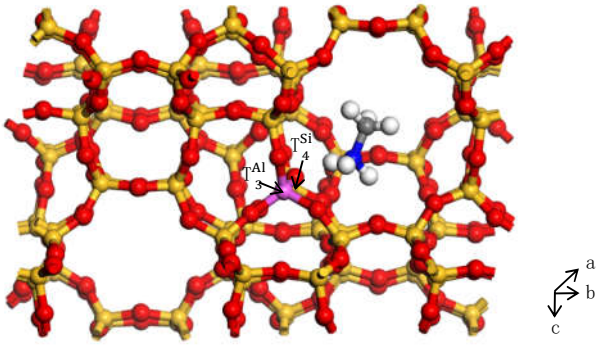


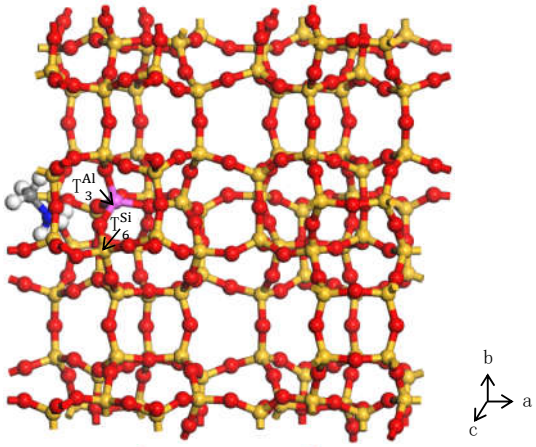
T^{Al}_{3}–0^{II}–T^{Si}_{2}



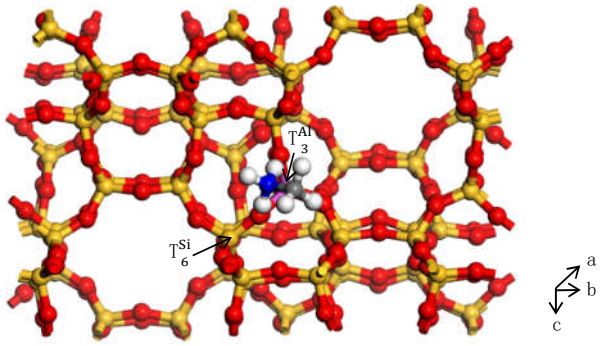


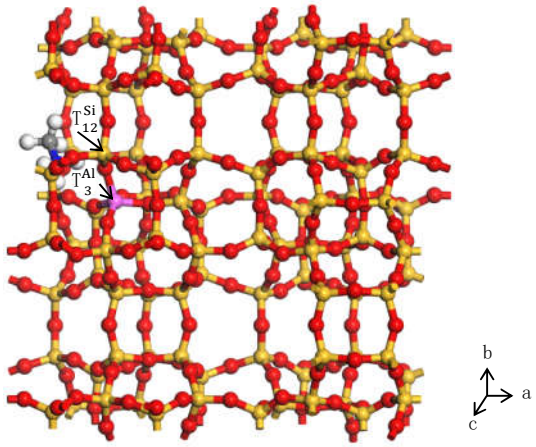
T^{Al}_{3}–0^{II}–T^{Si}_{4}



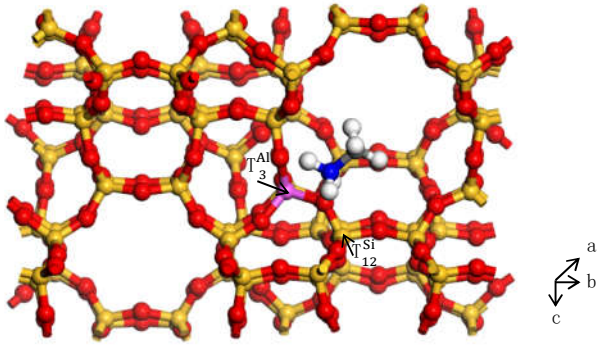


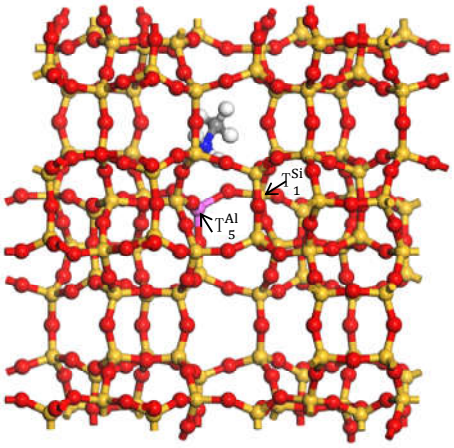
T^{Al}_{3}–0^{II}–T^{Si}_{6}



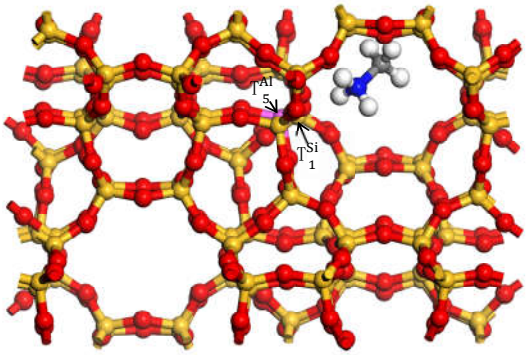


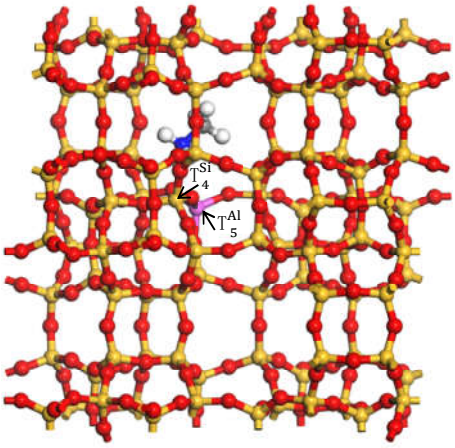
T^{Al}_{3}–0^{II}–T^{Si}_{12}



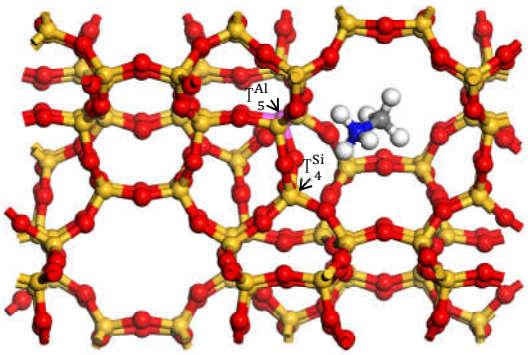


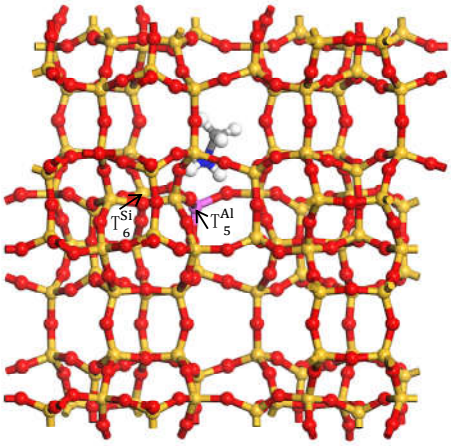
T^{Al}_{5}–0^{II}–T^{Si}_{1}



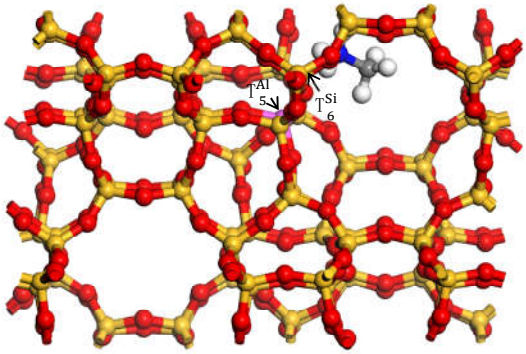


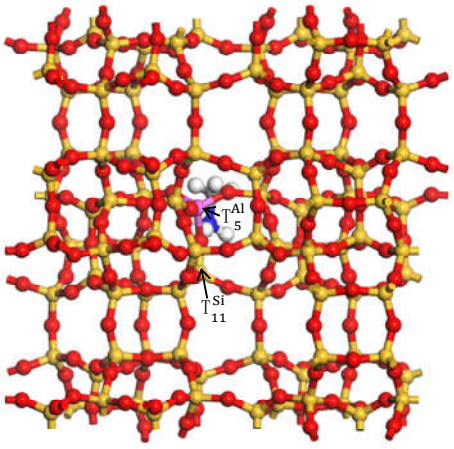
T^{Al}_{5}–0^{II}–T^{Si}_{4}



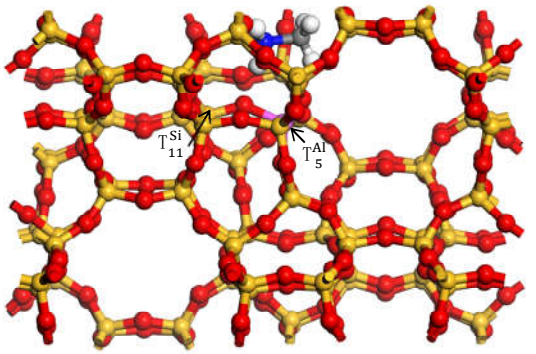


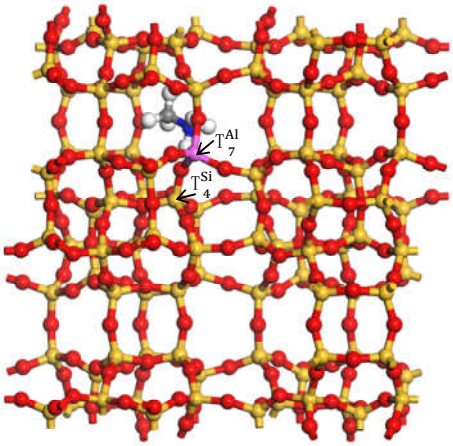
T^{Al}_{5}–0^{II}–T^{Si}_{6}



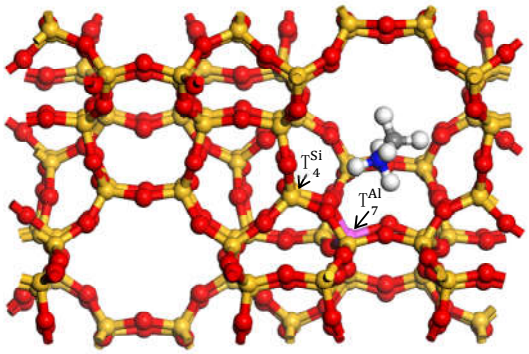


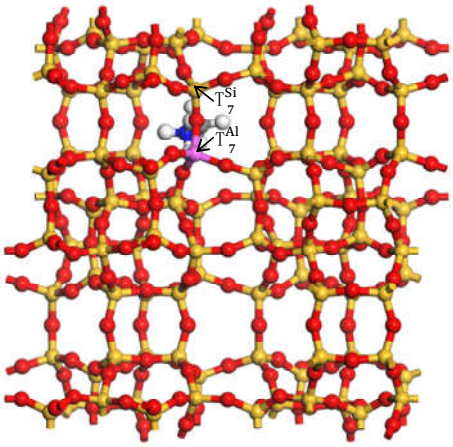
T^{Al}_{5}–0^{II}–T^{Si}_{11}



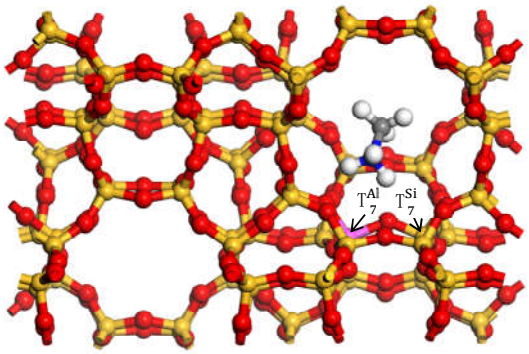


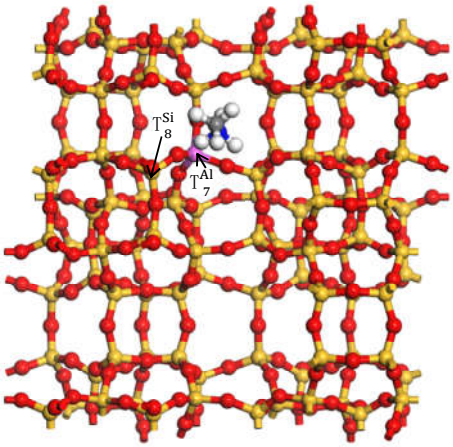
T^{Al}_{7}–0^{II}–T^{Si}_{4}



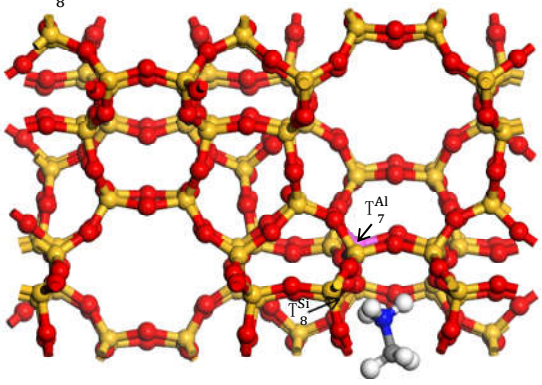


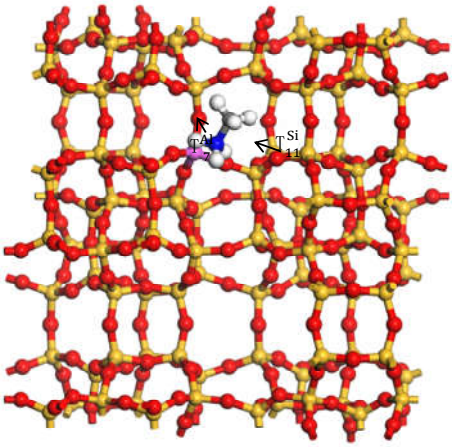
T^{Al}_{7}–0^{II}–T^{Si}_{7}



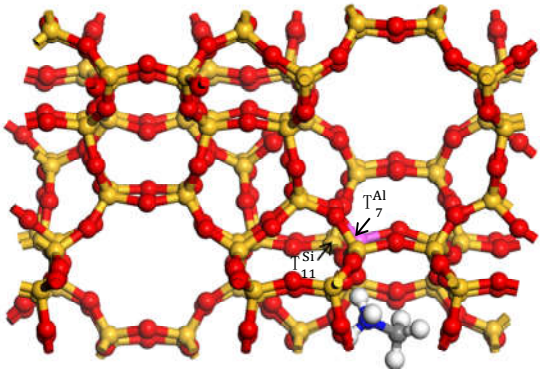


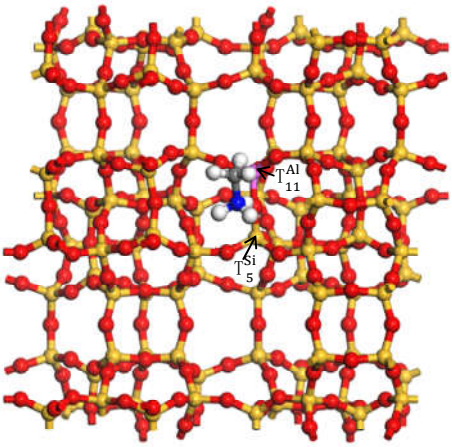
T^{Al}_{7}–0^{II}–T^{Si}_{8}



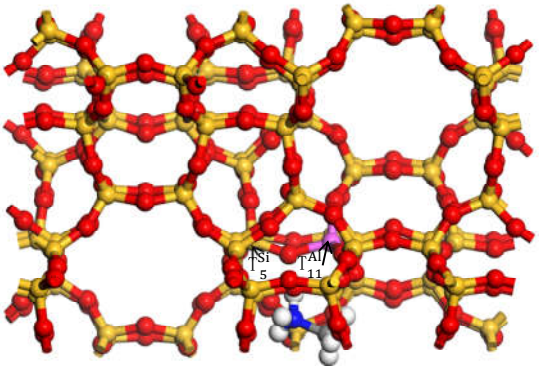


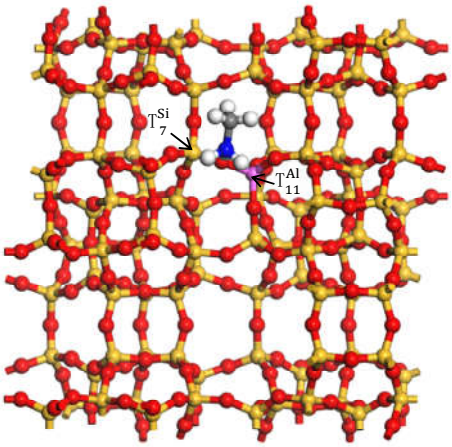
T^{Al}_{7}–0^{II}–T^{Si}_{11}



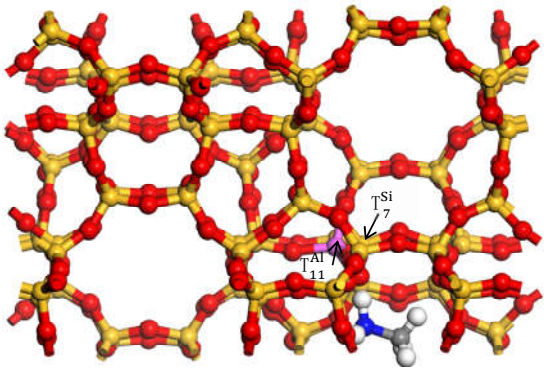


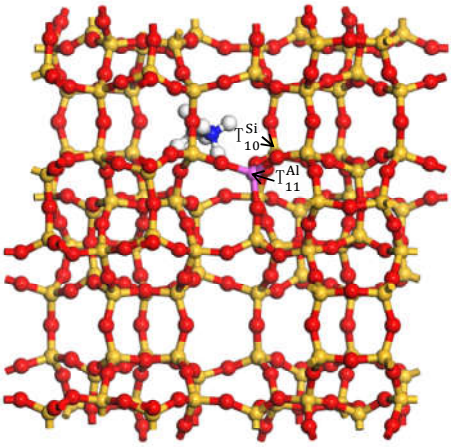
T^{Al}_{11}–O^{II}–T^{Si}_{5}



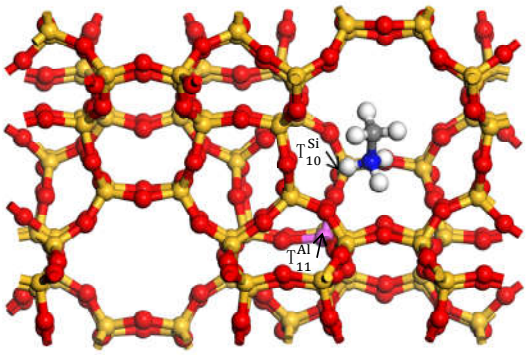


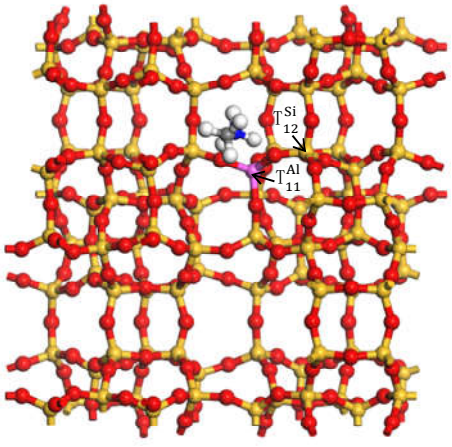
T^{Al}_{11}–O^{II}–T^{Si}_{7}



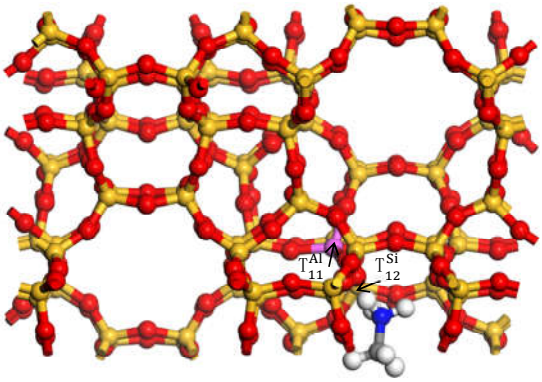


T^{Al}_{11}–O^{II}–T^{Si}_{10}





T^{Al}_{11}–O^{II}–T^{Si}_{12}



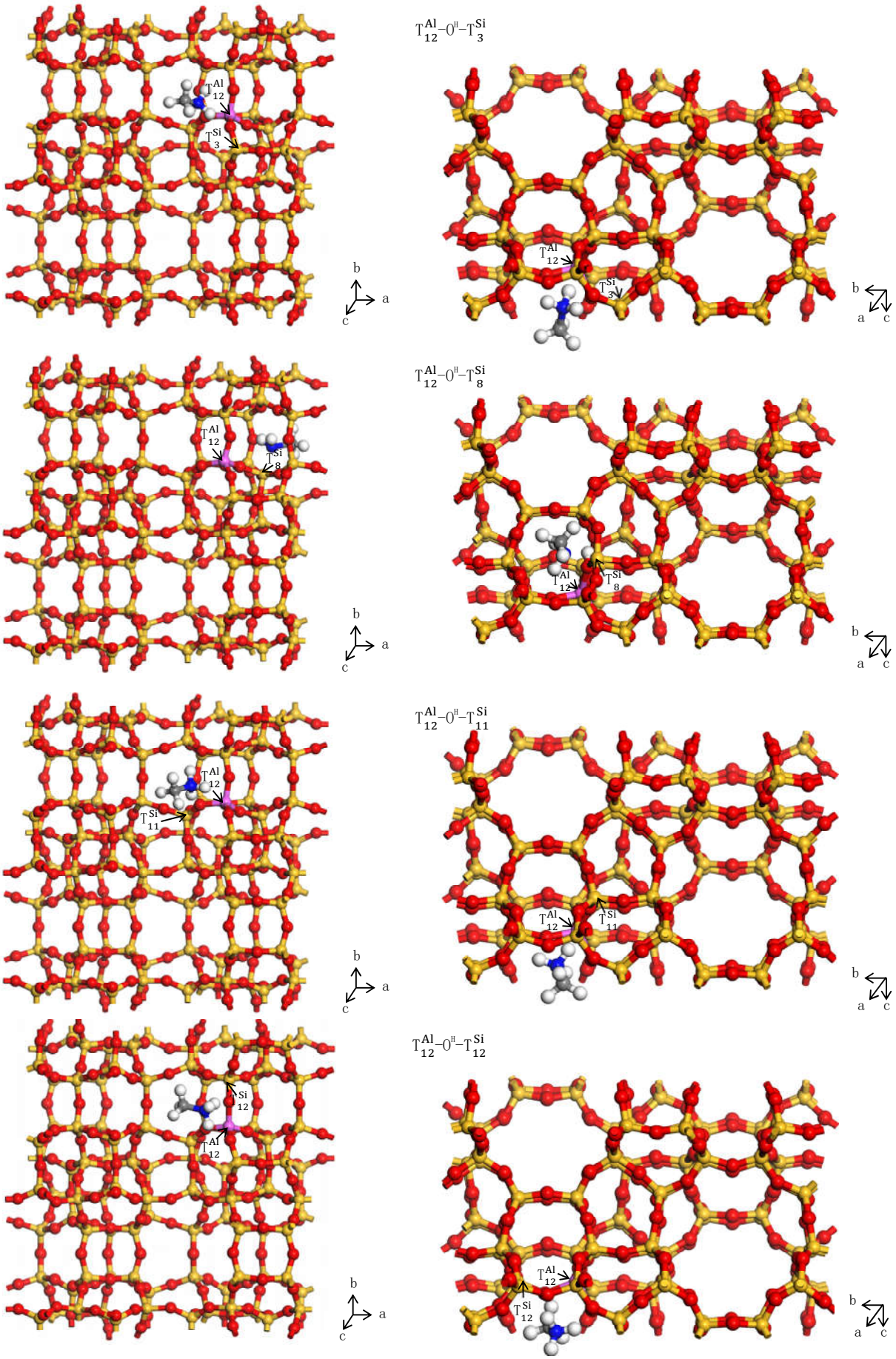
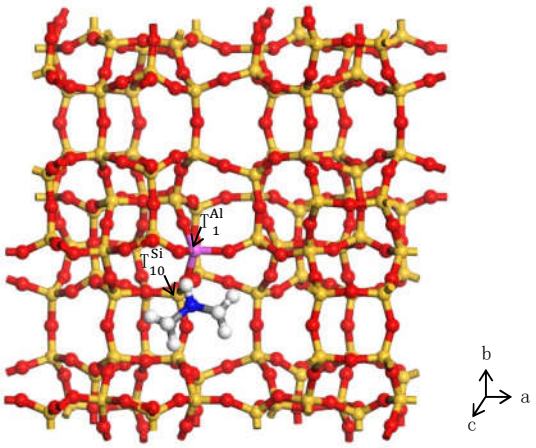
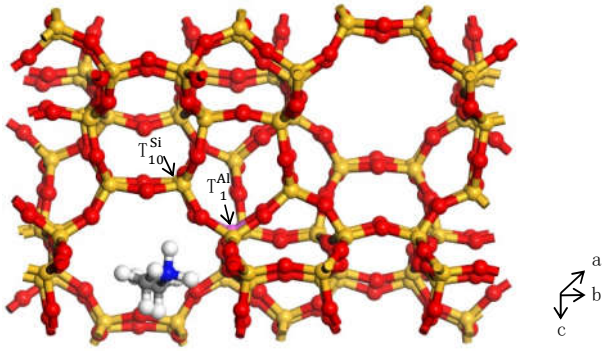
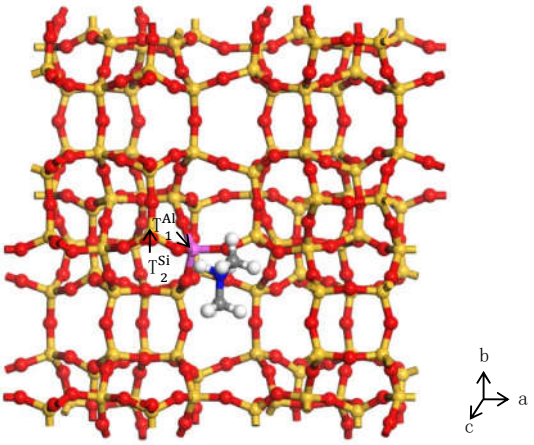


Figure S5 The optimized configurations of the adsorption of NH_2CH_3 at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework

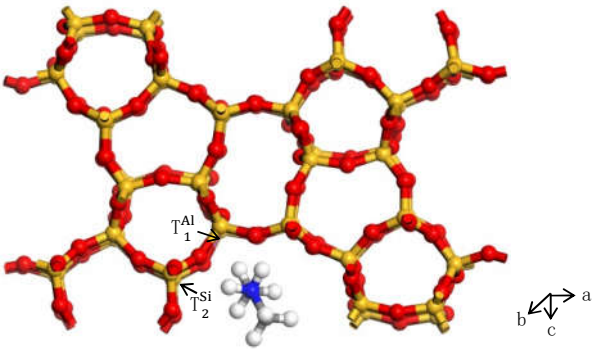


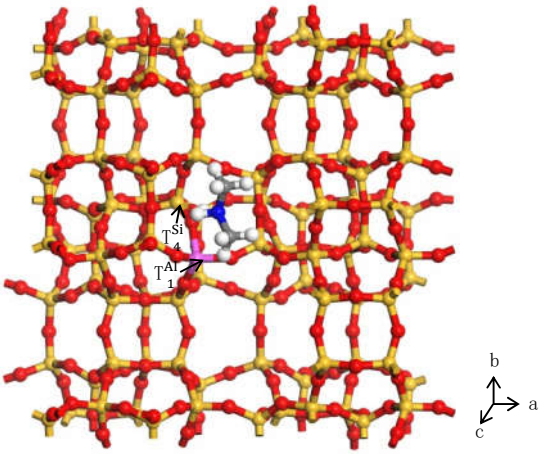
T^{Al}_{1}–O^{H}–T^{Si}_{10}



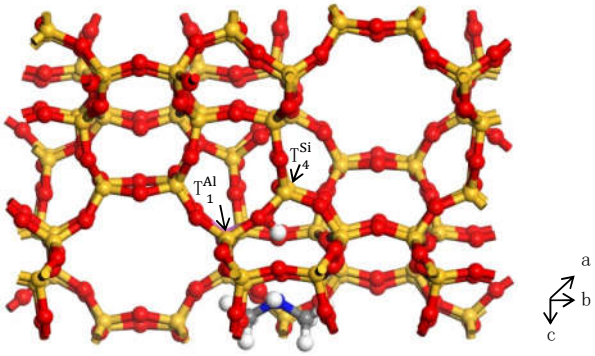


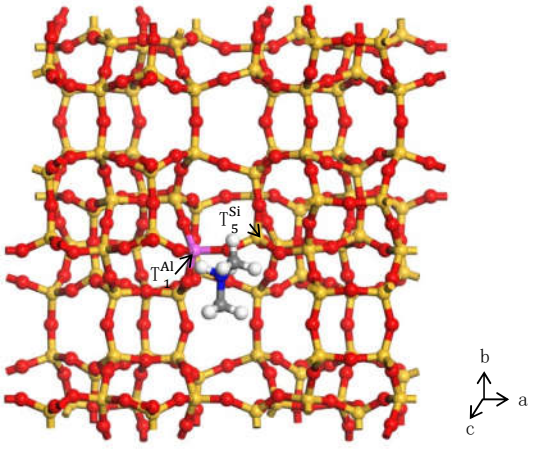
T^{Al}_{1}–O^{H}–T^{Si}_{2}



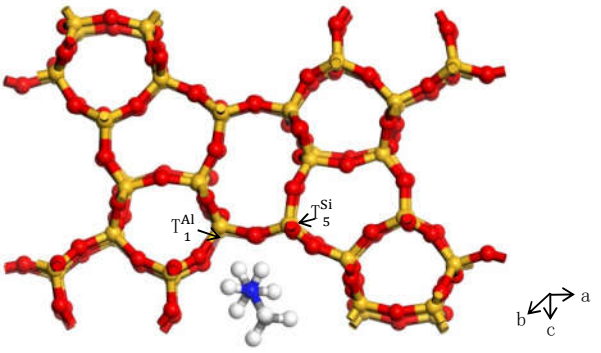


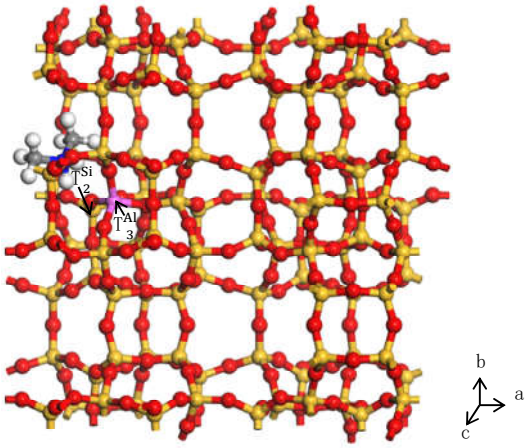
T^{Al}_{1}–O^{H}–T^{Si}_{4}



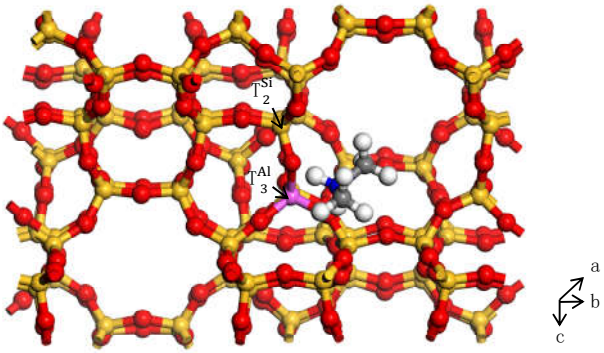


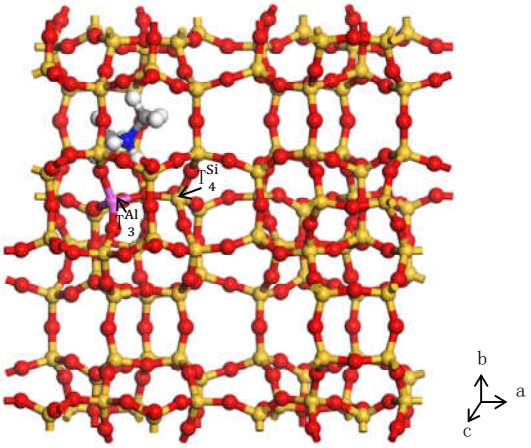
T^{Al}_{1}–O^{H}–T^{Si}_{5}



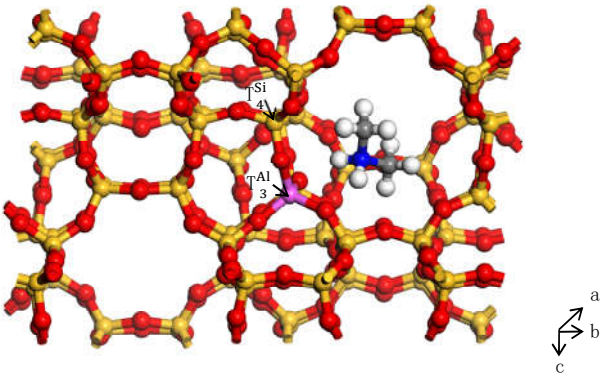


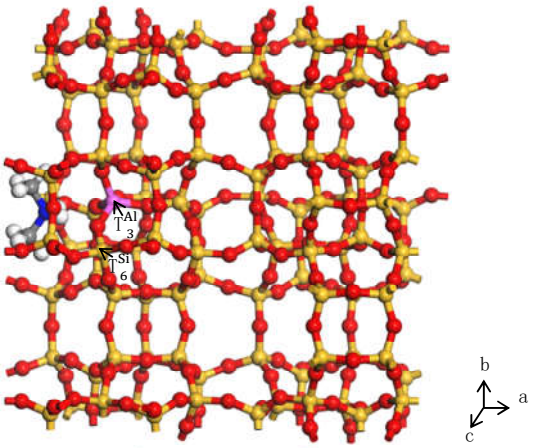
T^{Al}_{3}–O^{II}–T^{Si}_{2}



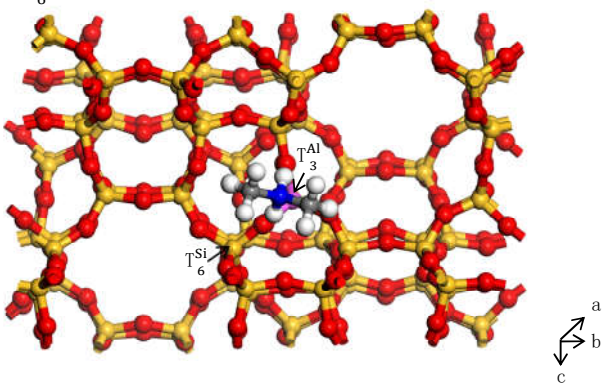


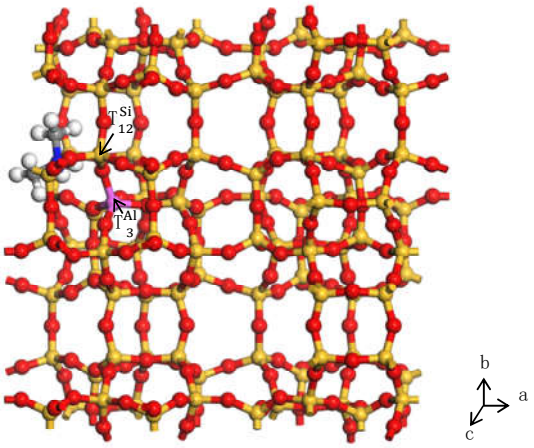
T^{Al}_{3}–O^{II}–T^{Si}_{4}



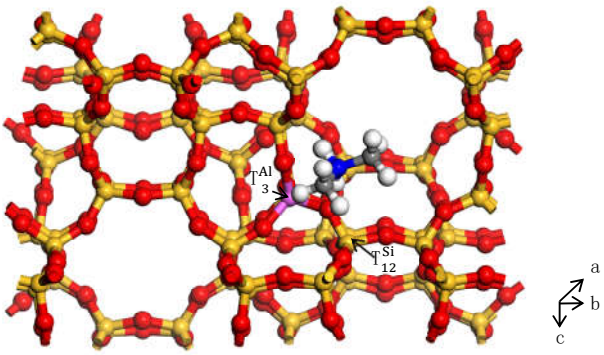


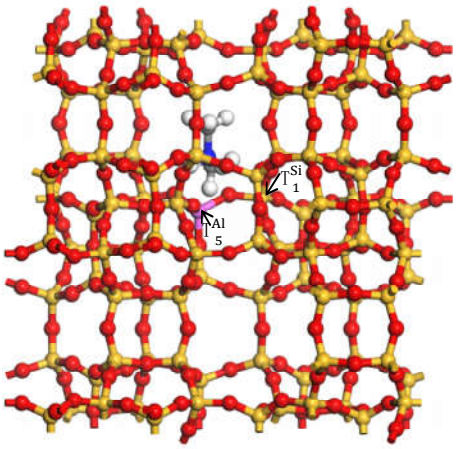
T^{Al}_{3}–O^{II}–T^{Si}_{6}



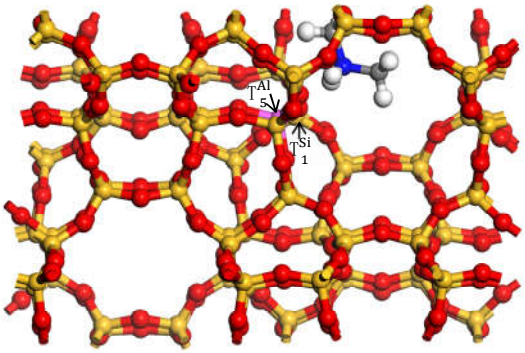


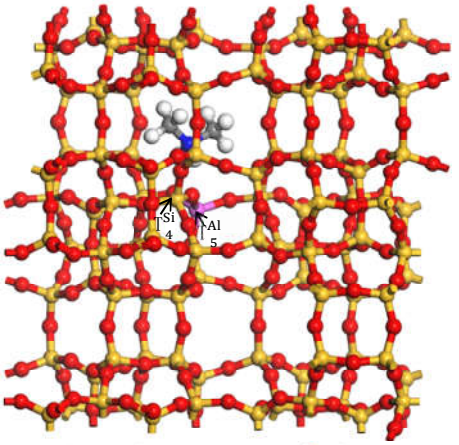
T^{Al}_{3}–O^{II}–T^{Si}_{12}



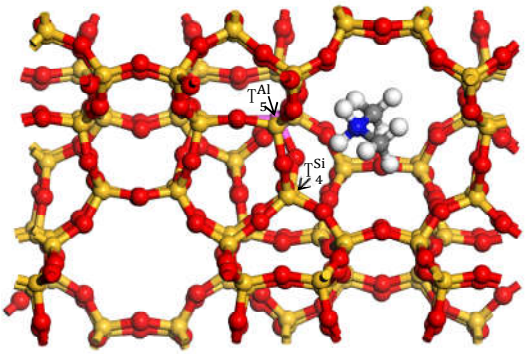


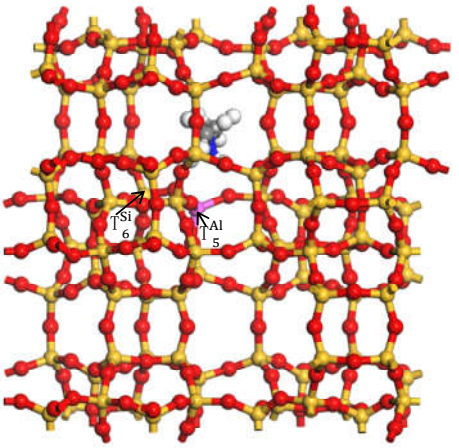
T^{Al}_{5}–O^{H}–T^{Si}_{1}



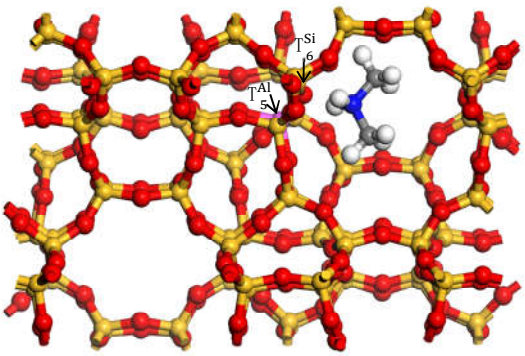


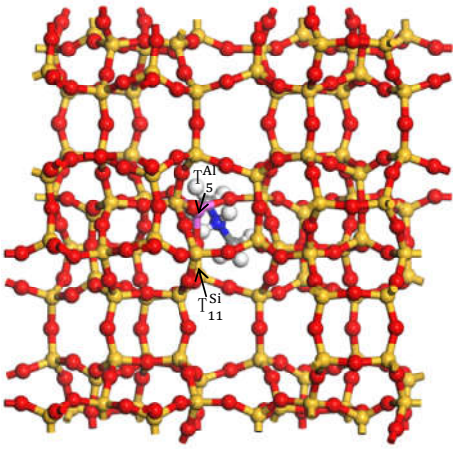
T^{Al}_{5}–O^{H}–T^{Si}_{4}



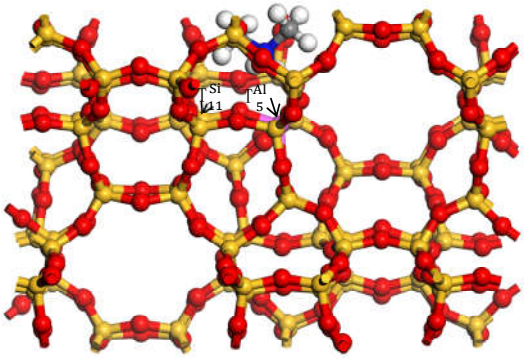


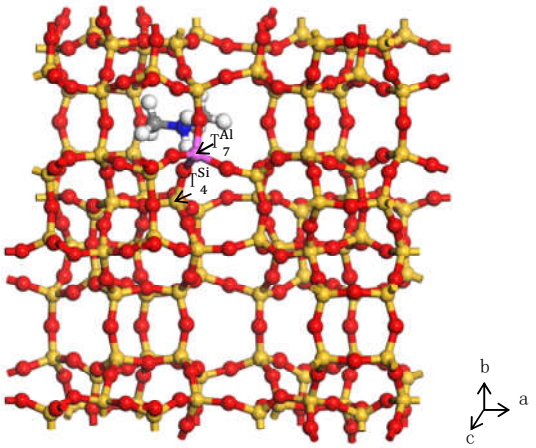
T^{Al}_{5}–O^{H}–T^{Si}_{6}



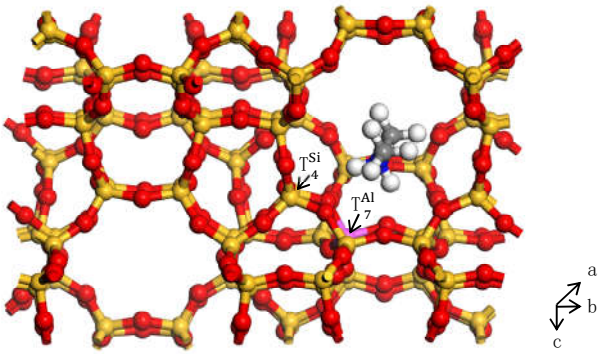


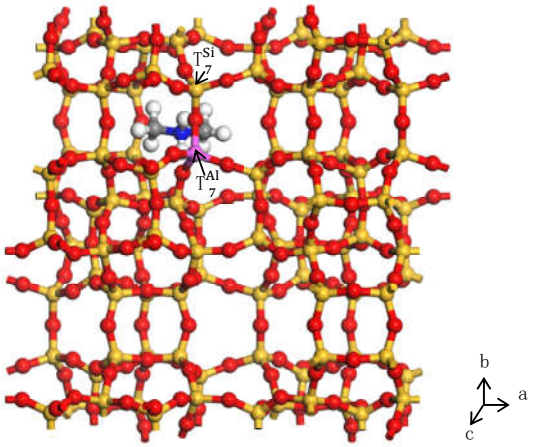
T^{Al}_{5}–O^{H}–T^{Si}_{11}



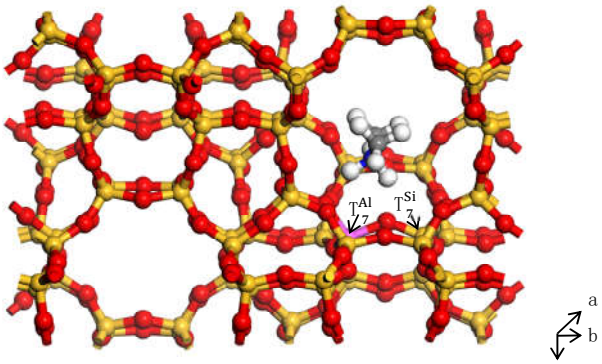


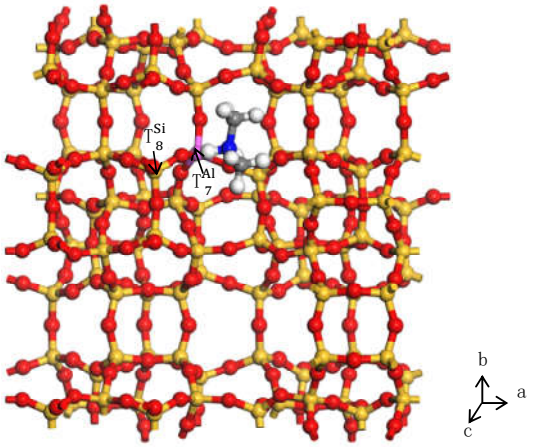
T^{Al}_{7}–O^{H}–T^{Si}_{4}



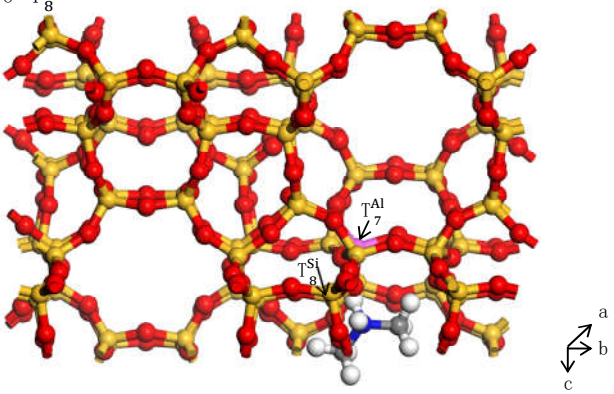


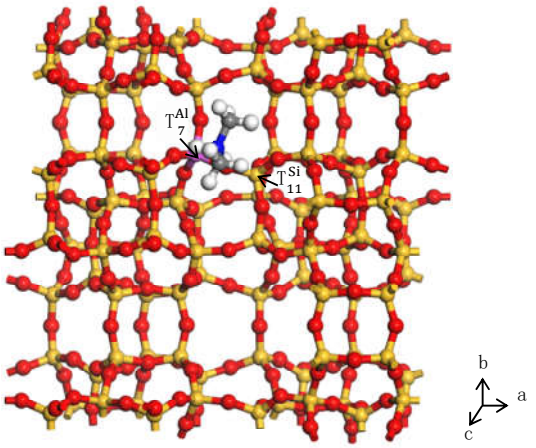
T^{Al}_{7}–O^{H}–T^{Si}_{7}



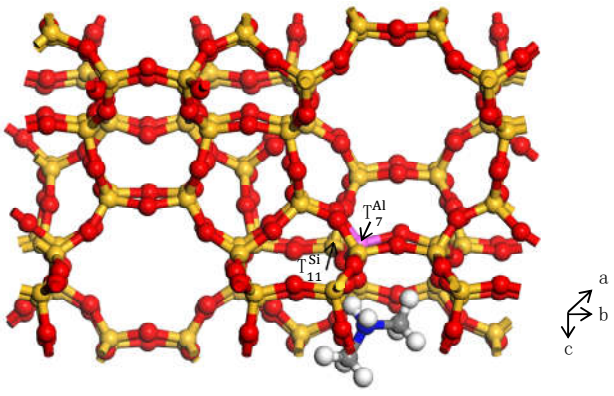


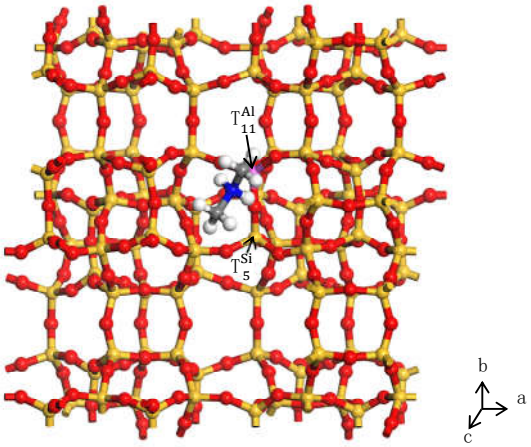
T^{Al}_{7}–O^{H}–T^{Si}_{8}



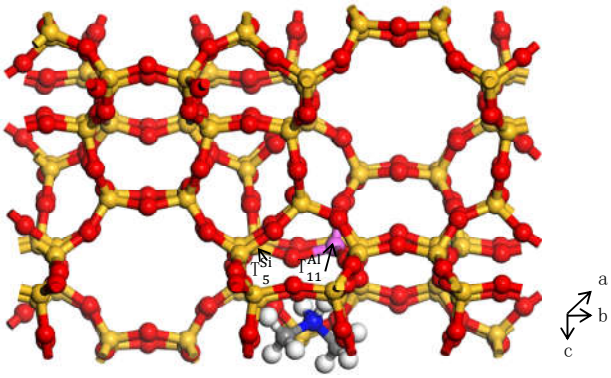


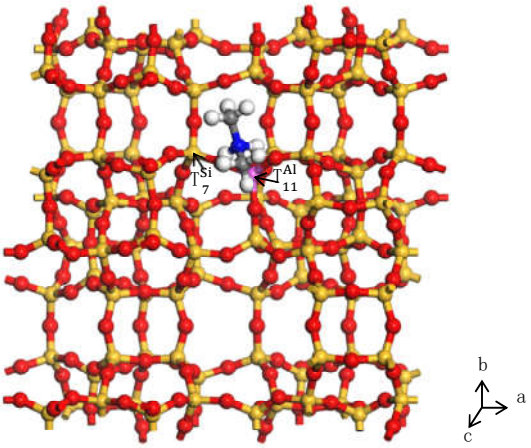
T^{Al}_{7}–O^{H}–T^{Si}_{11}



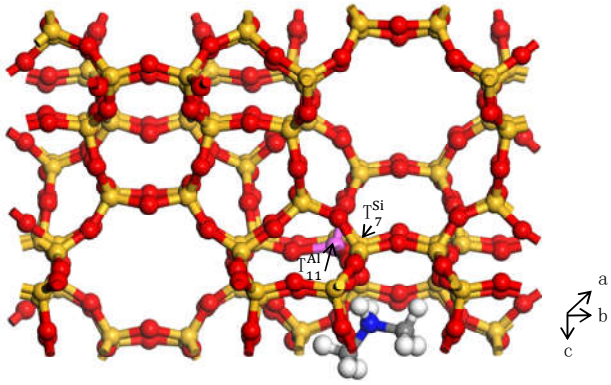


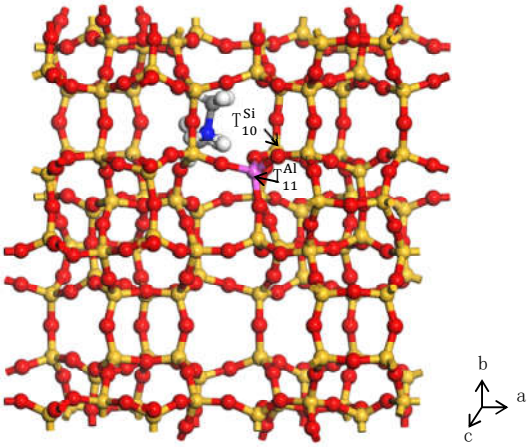
T^{Al}_{11}–O^{II}–T^{Si}_{5}



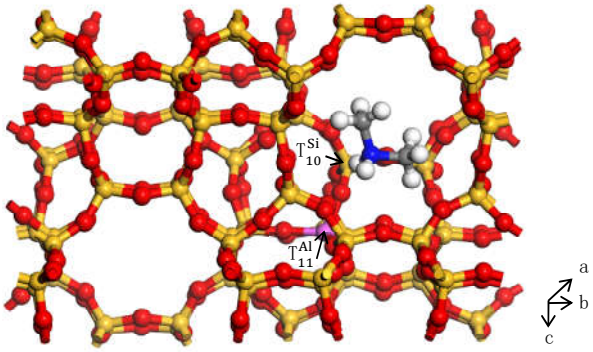


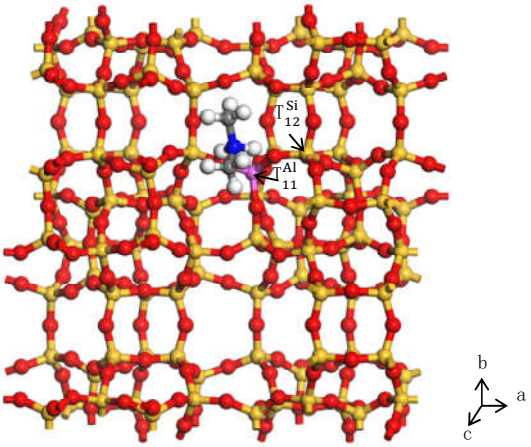
T^{Al}_{11}–O^{II}–T^{Si}_{7}



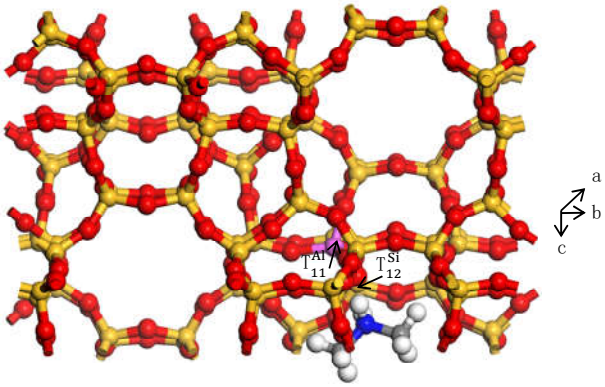


T^{Al}_{11}–O^{II}–T^{Si}_{10}





T^{Al}_{11}–O^{II}–T^{Si}_{12}



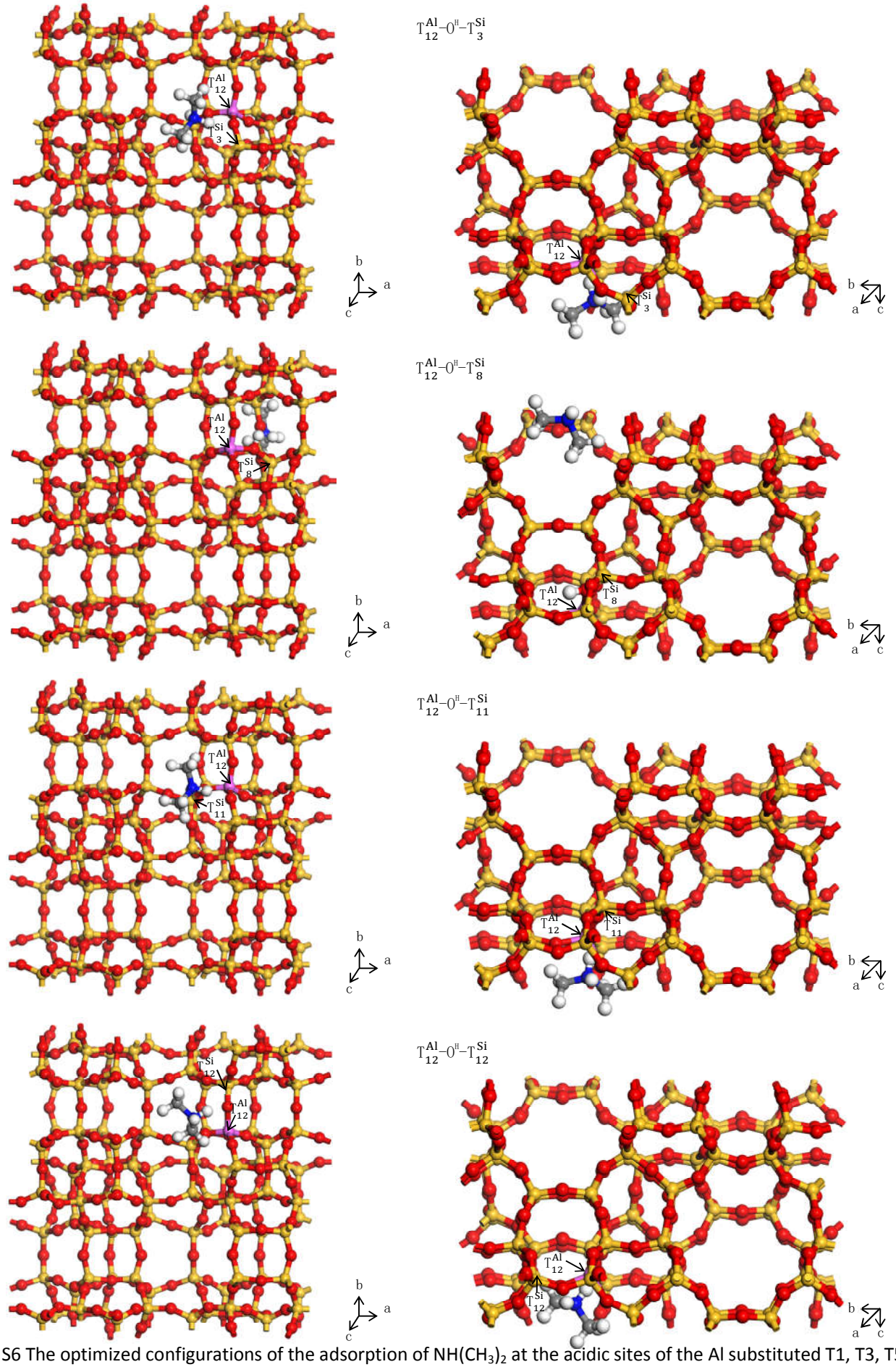
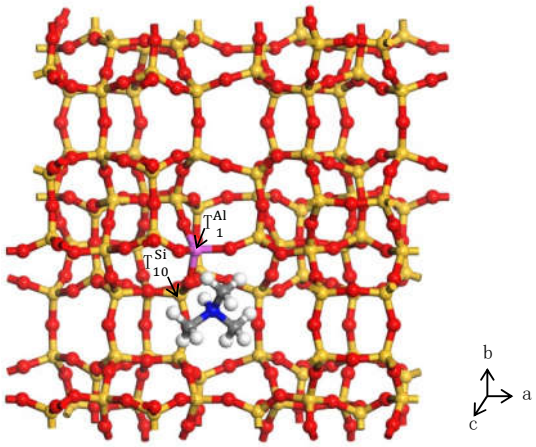
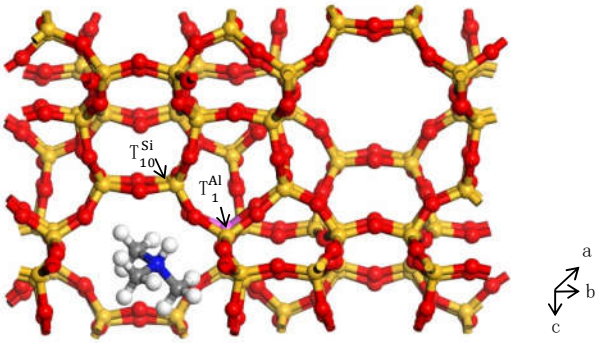
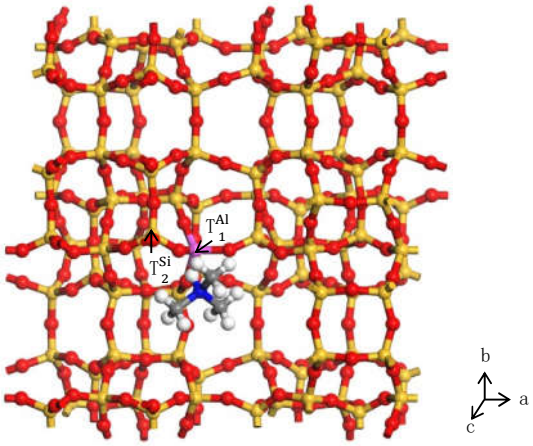


Figure S6 The optimized configurations of the adsorption of $NH(CH_3)_2$ at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework

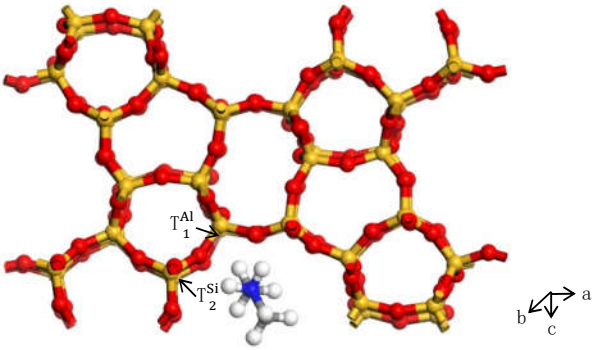


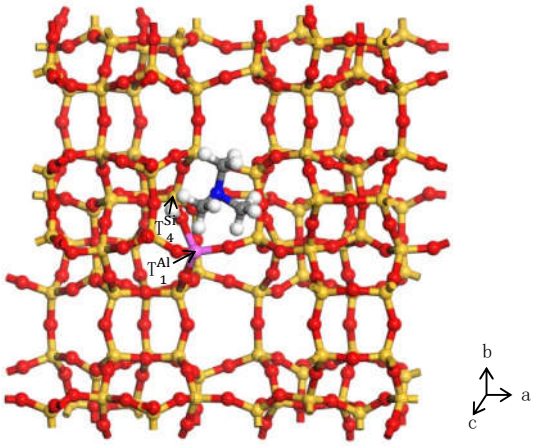
T^{Al}_{1}–O^{II}–T^{Si}_{10}



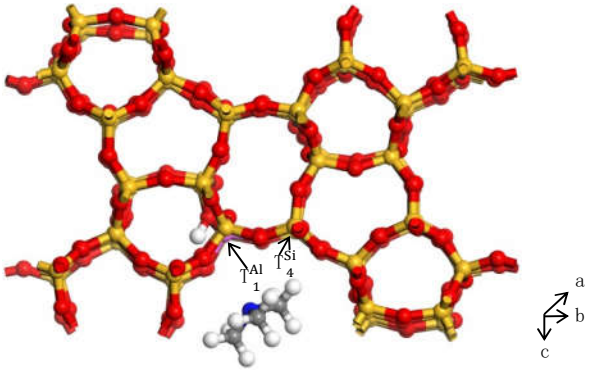


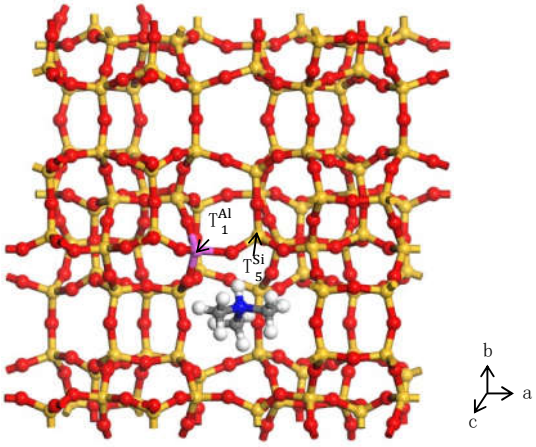
T^{Al}_{1}–O^{II}–T^{Si}_{2}



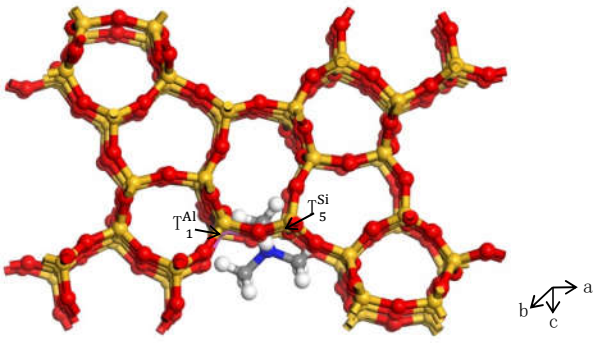


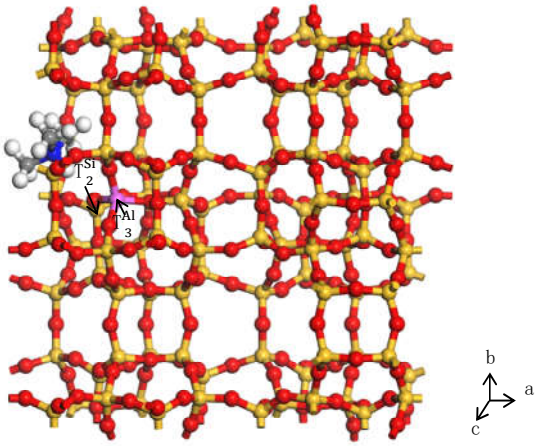
T^{Al}_{1}–O^{II}–T^{Si}_{4}



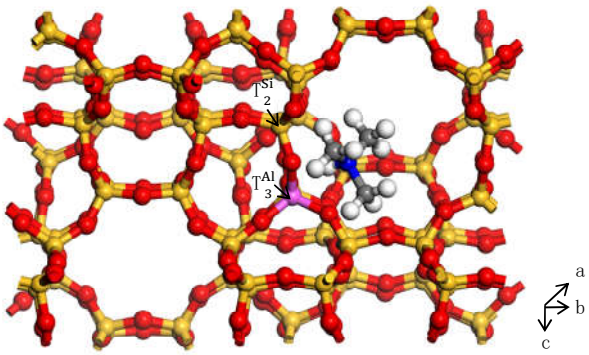


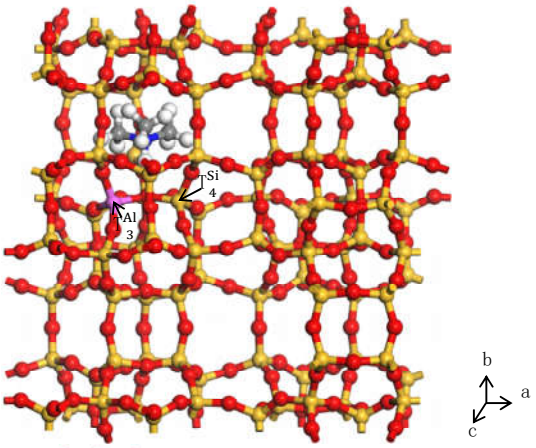
T^{Al}_{1}–O^{II}–T^{Si}_{5}



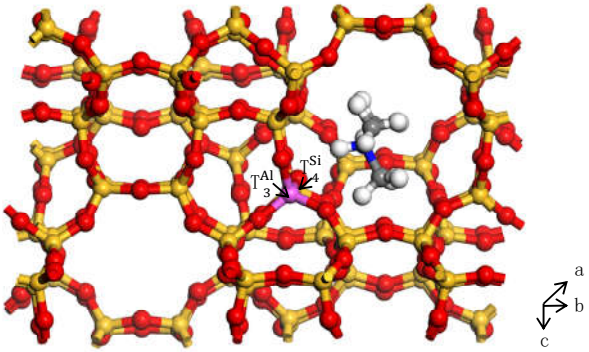


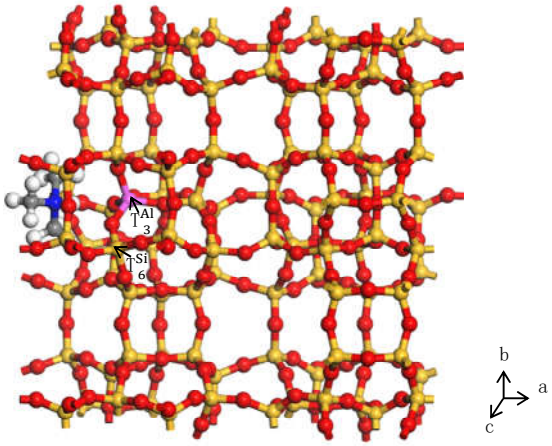
T^{Al}_{3}–O^{H}–T^{Si}_{2}



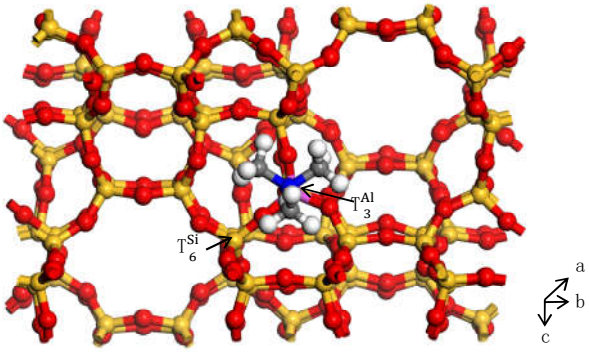


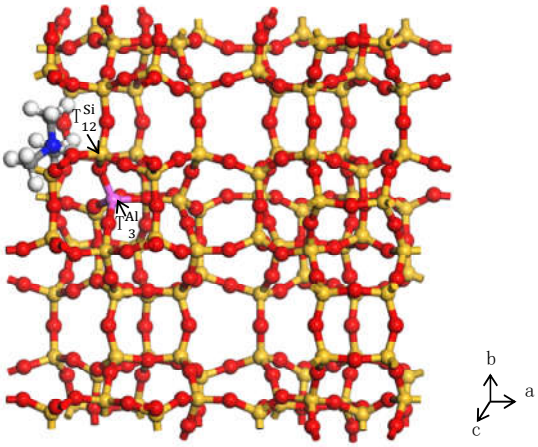
T^{Al}_{3}–O^{H}–T^{Si}_{4}



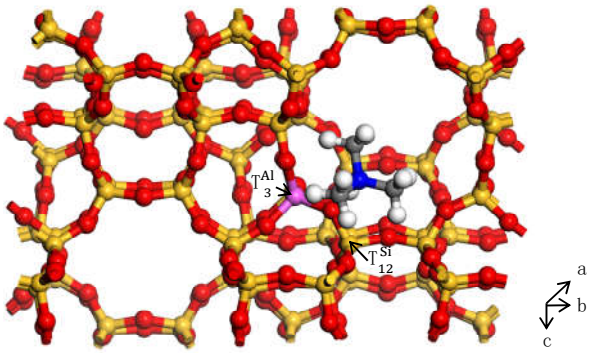


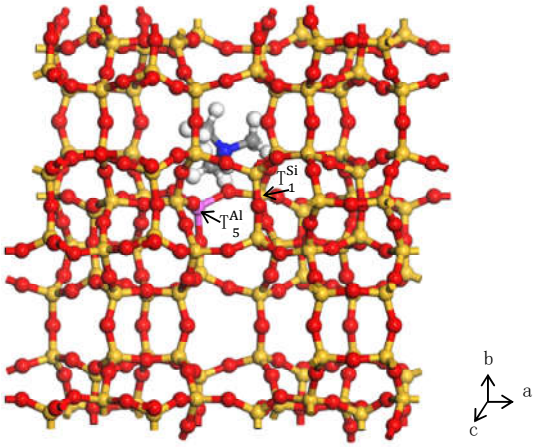
T^{Al}_{3}–O^{H}–T^{Si}_{6}



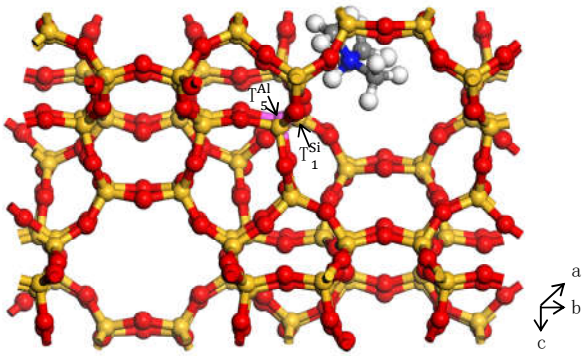


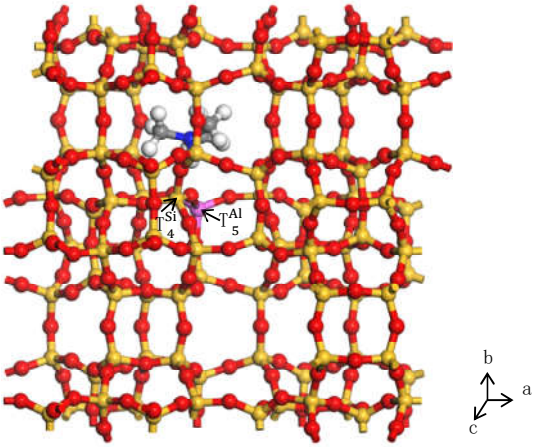
T^{Al}_{3}–O^{H}–T^{Si}_{12}



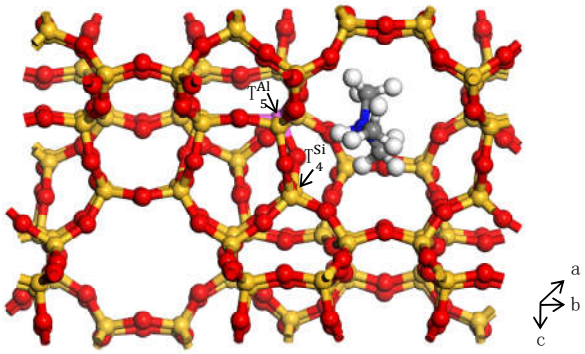


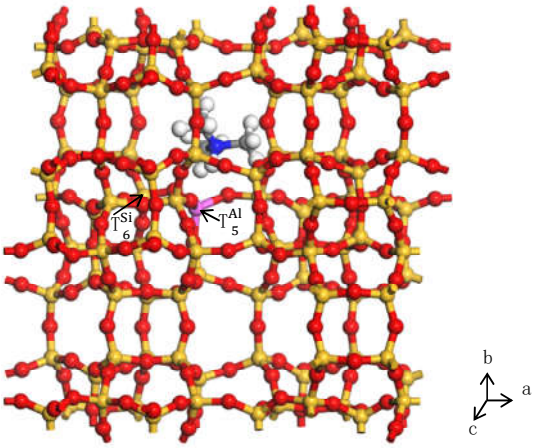
T^{Al}_{5}–O^{II}–T^{Si}_{1}



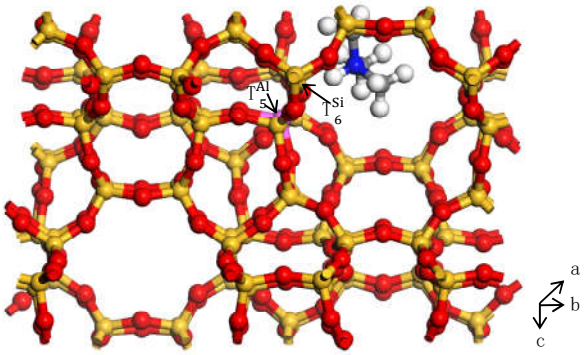


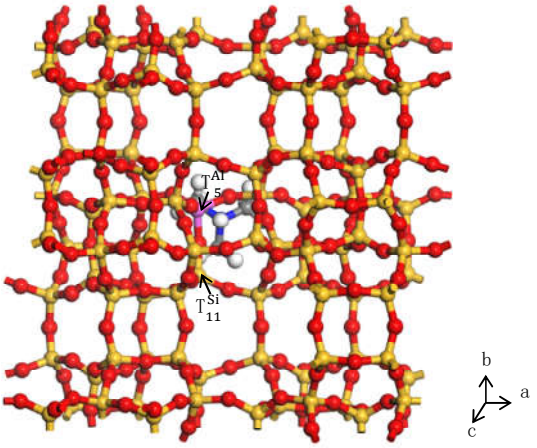
T^{Al}_{5}–O^{II}–T^{Si}_{4}



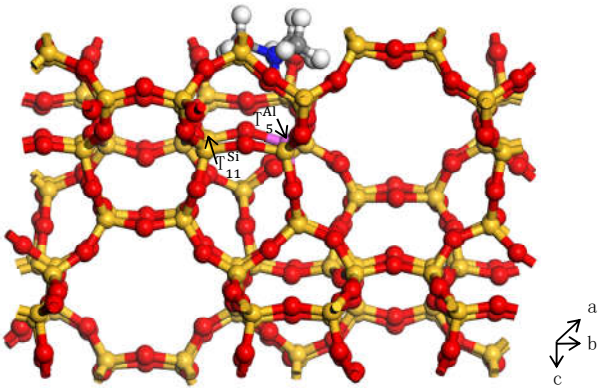


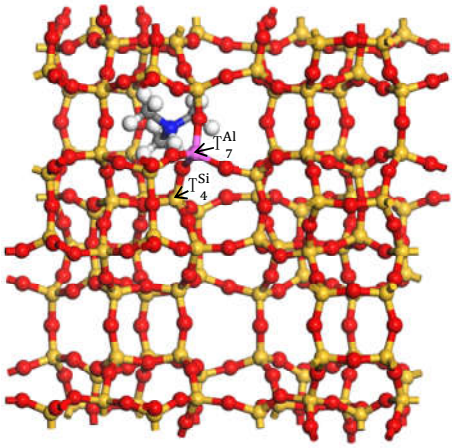
T^{Al}_{5}–O^{II}–T^{Si}_{6}



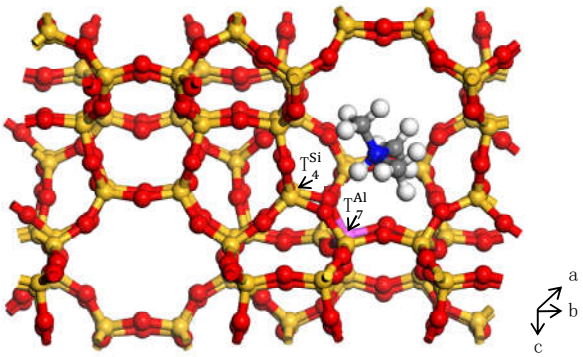


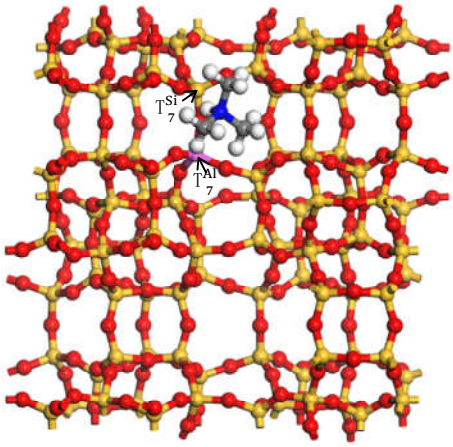
T^{Al}_{5}–O^{II}–T^{Si}_{11}



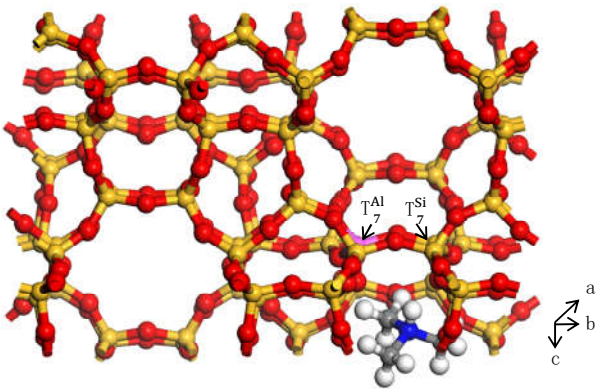


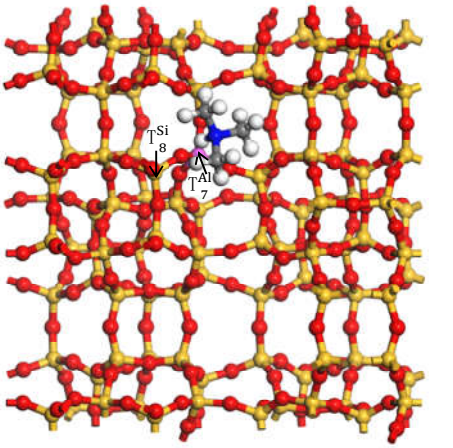
T^{Al}_{7}–O^{H}–T^{Si}_{4}



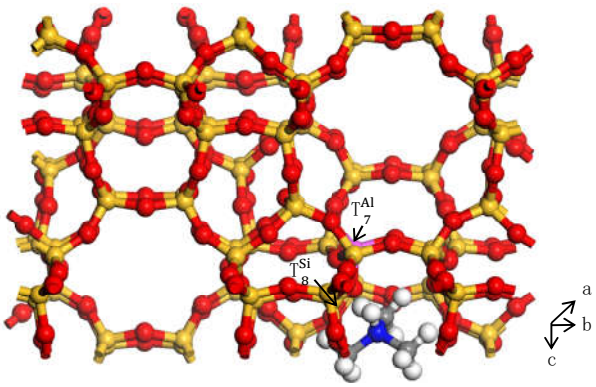


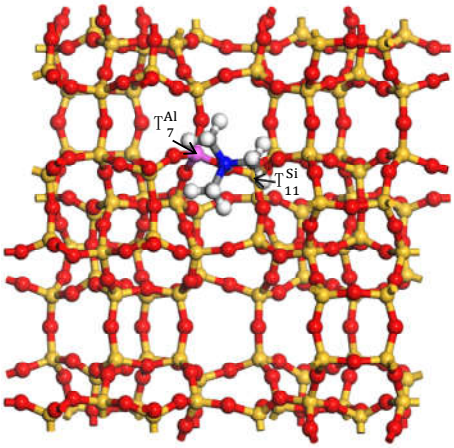
T^{Al}_{7}–O^{H}–T^{Si}_{7}



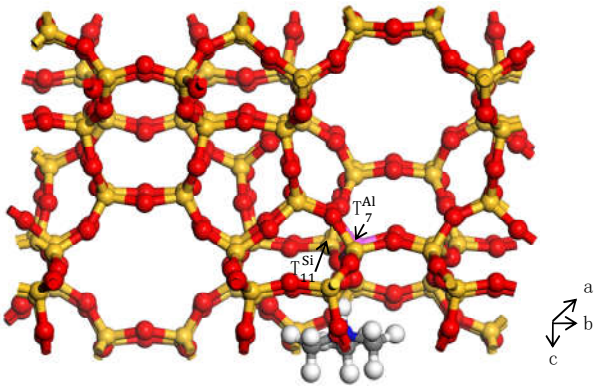


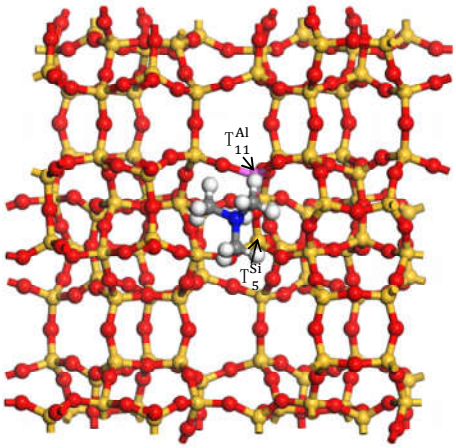
T^{Al}_{7}–O^{H}–T^{Si}_{8}



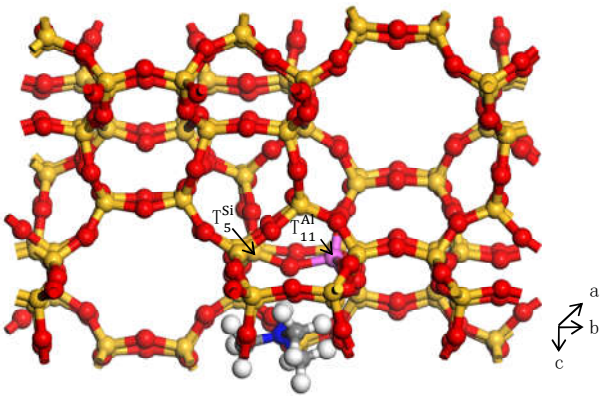


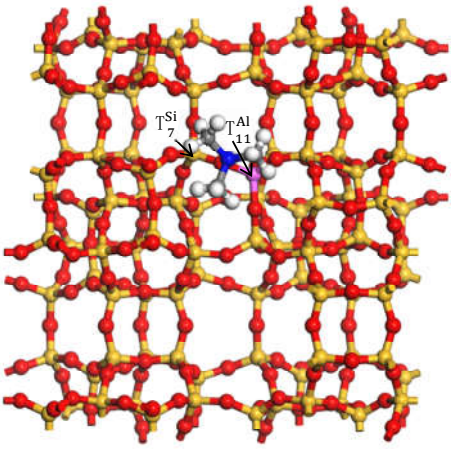
T^{Al}_{7}–O^{H}–T^{Si}_{11}



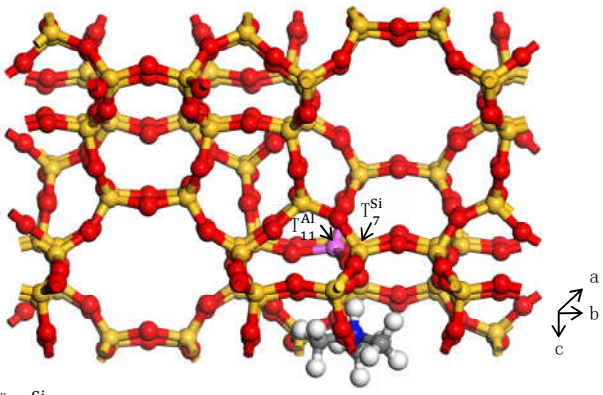


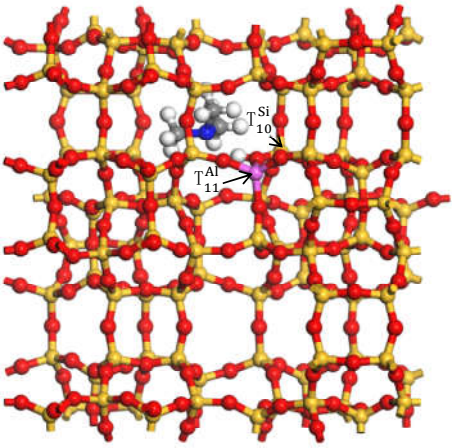
T^{Al}_{11}–O^{II}–T^{Si}_{5}



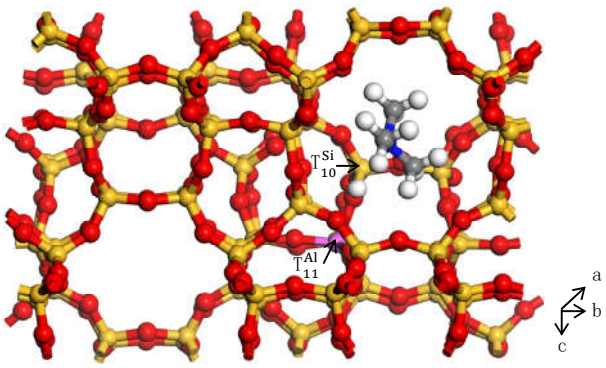


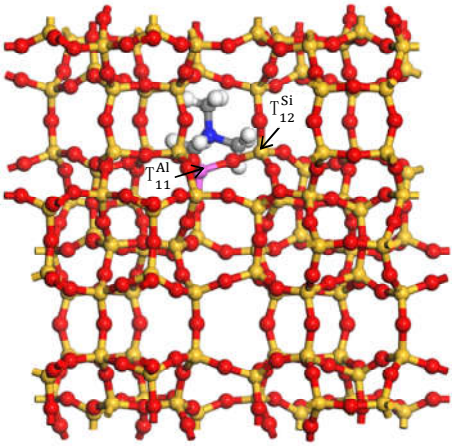
T^{Al}_{11}–O^{II}–T^{Si}_{7}



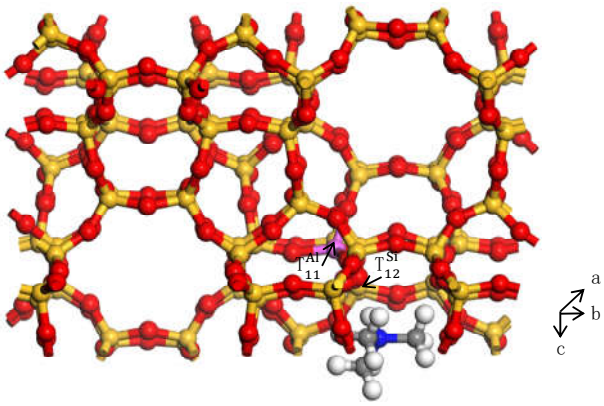


T^{Al}_{11}–O^{II}–T^{Si}_{10}





T^{Al}_{11}–O^{II}–T^{Si}_{12}



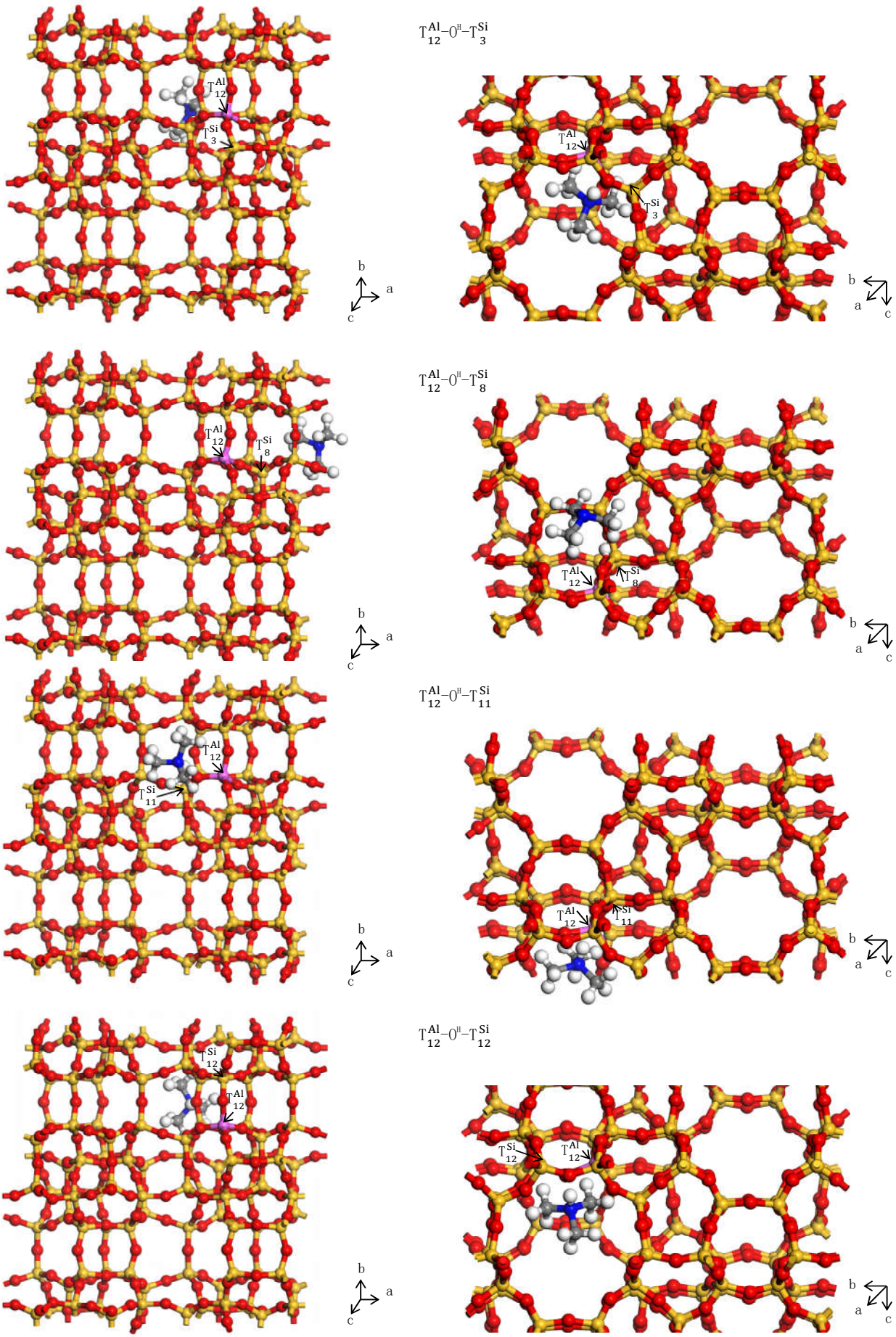
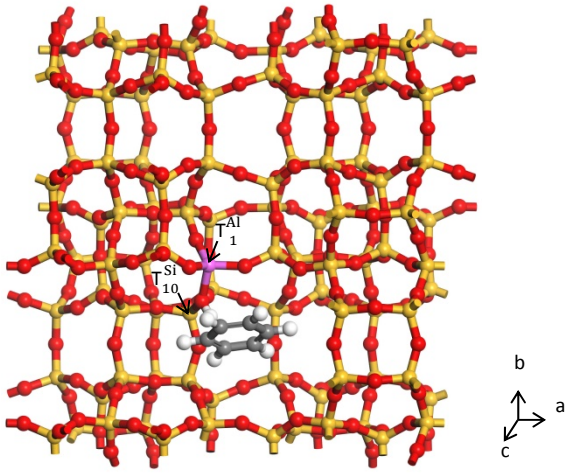
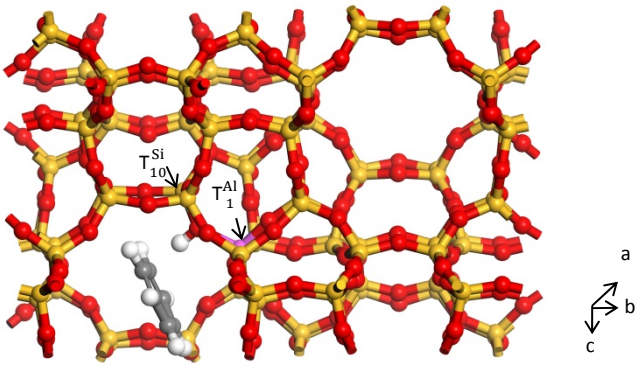
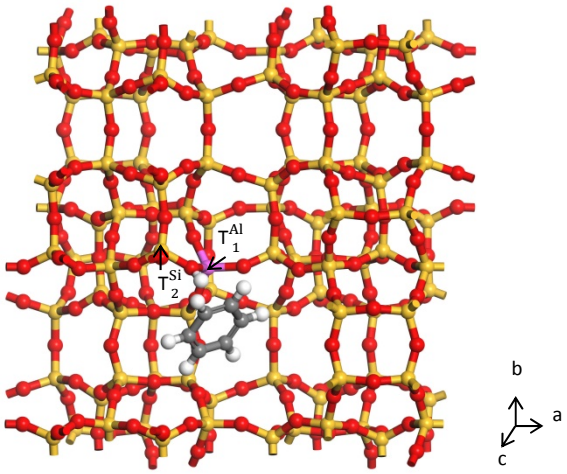


Figure S7 The optimized configurations of the adsorption of $N(CH_3)_3$ at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework

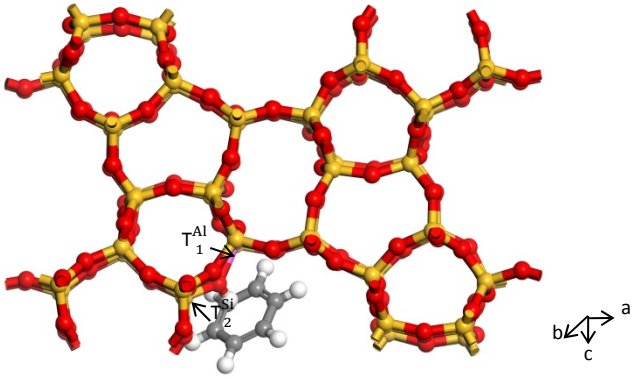


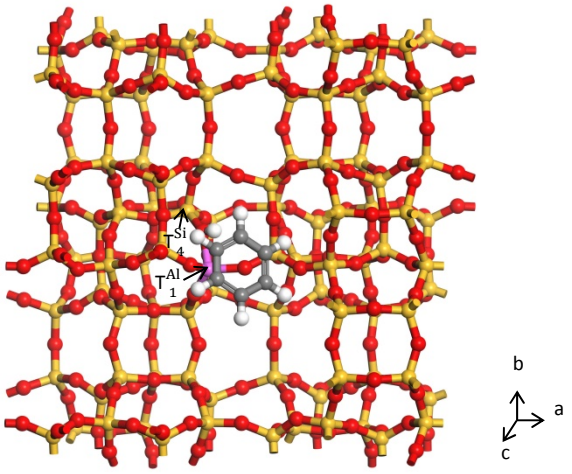
T^{Al}_{1}-O^{H}-T^{Si}_{10}



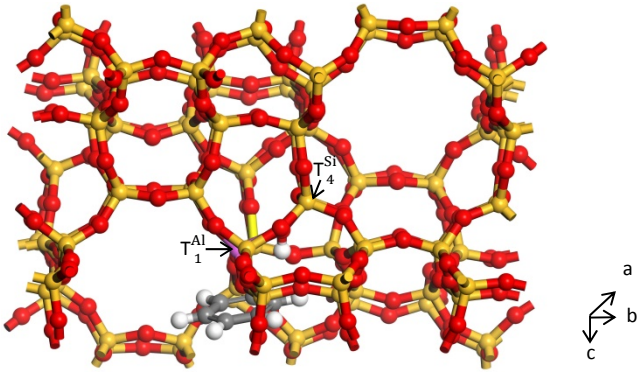


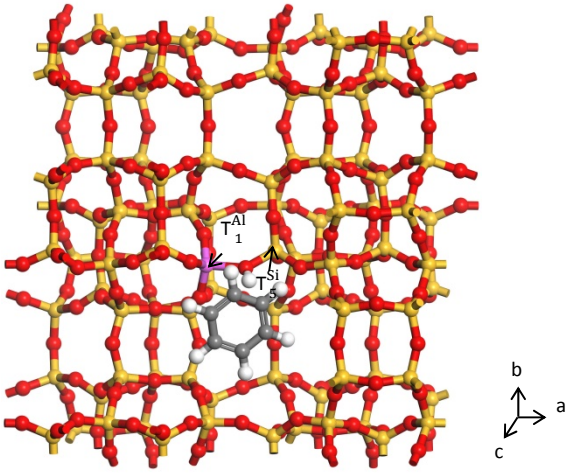
T^{Al}_{1}-O^{H}-T^{Si}_{2}



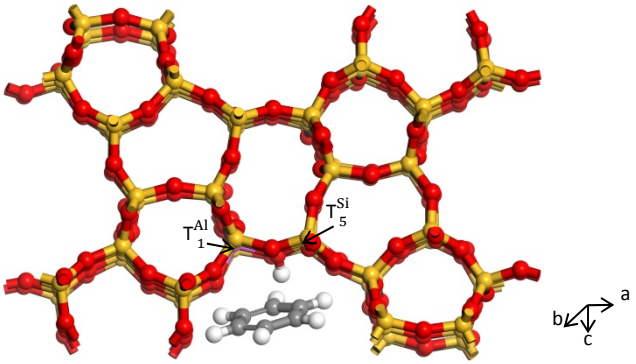


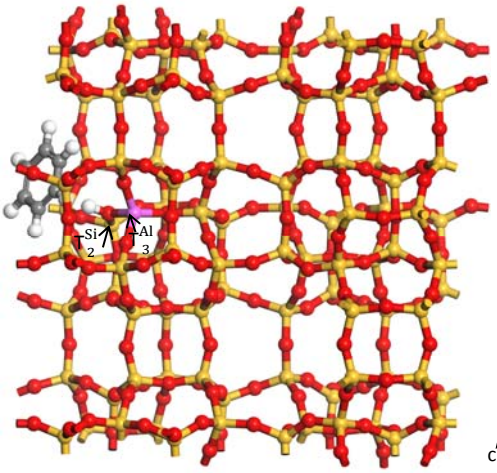
T^{Al}_{1}-O^{H}-T^{Si}_{4}



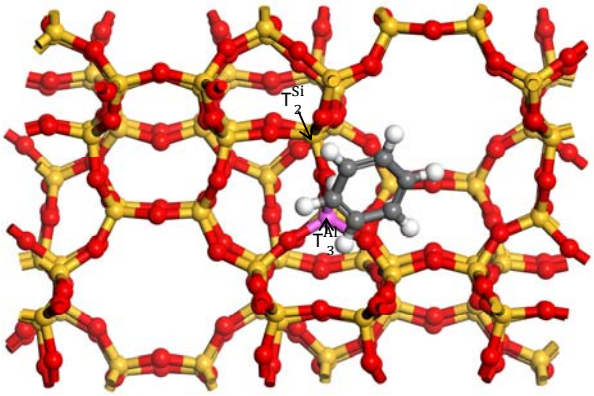


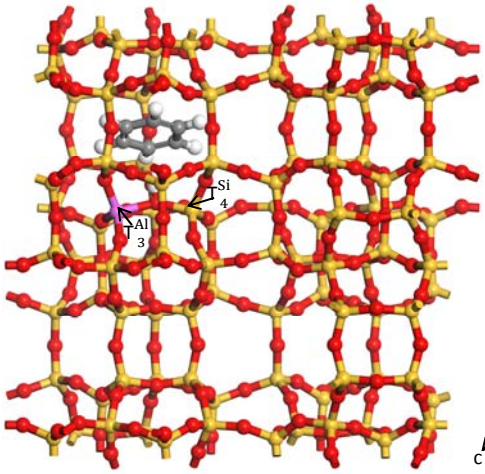
T^{Al}_{1}-O^{H}-T^{Si}_{5}



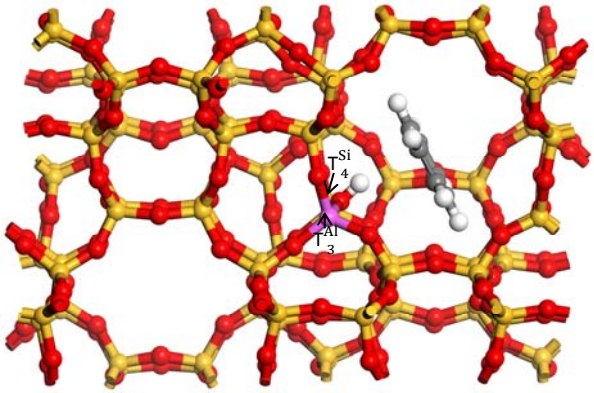


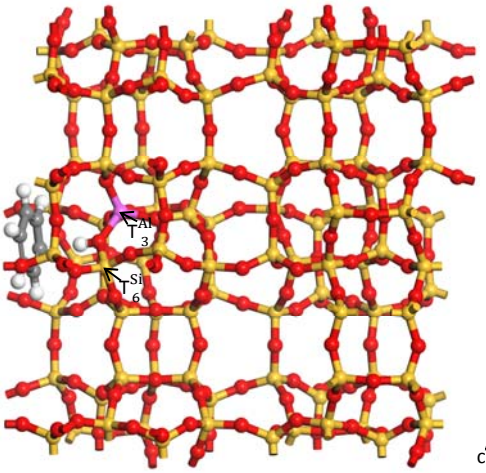
T^{Al}_{3}-O^{H}-T^{Si}_{2}



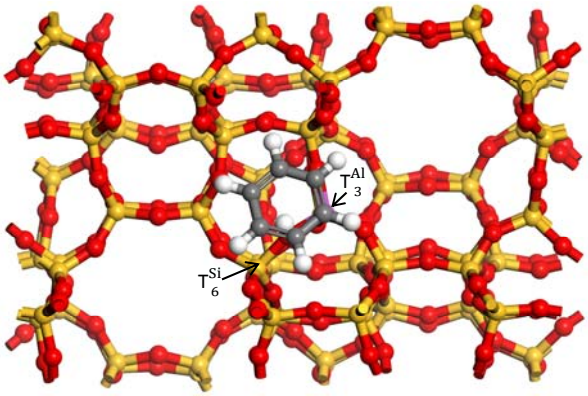


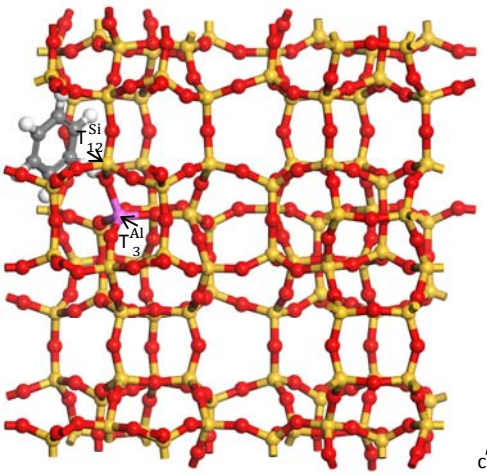
T^{Al}_{3}-O^{H}-T^{Si}_{4}



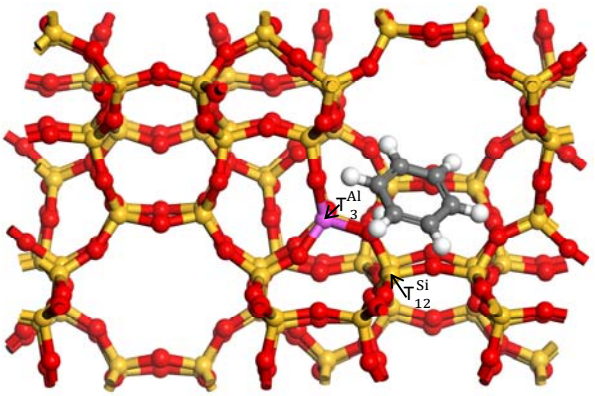


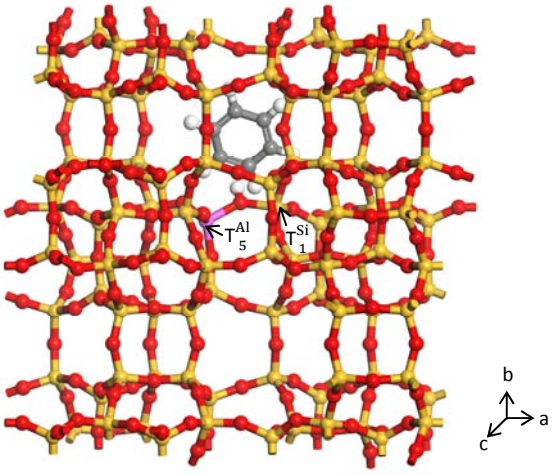
T^{Al}_{3}-O^{H}-T^{Si}_{6}



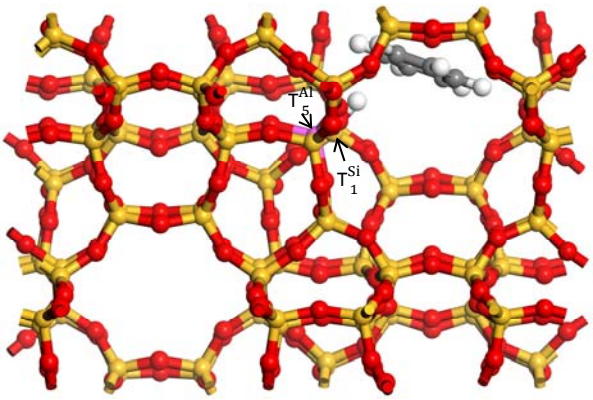


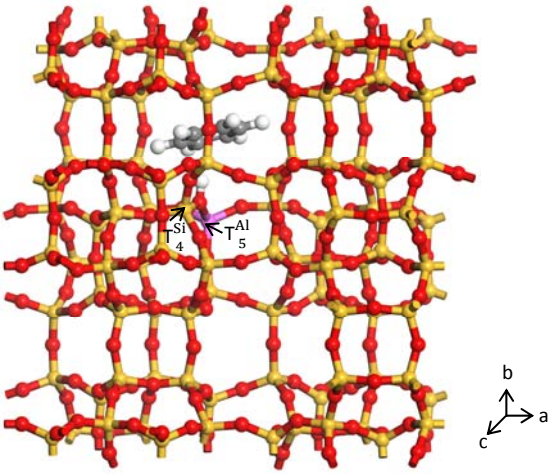
T^{Al}_{3}-O^{H}-T^{Si}_{12}



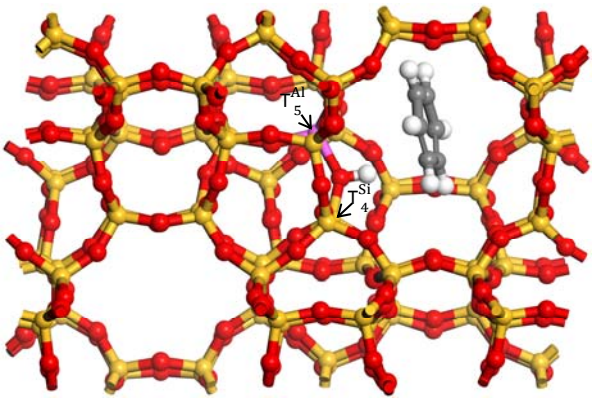


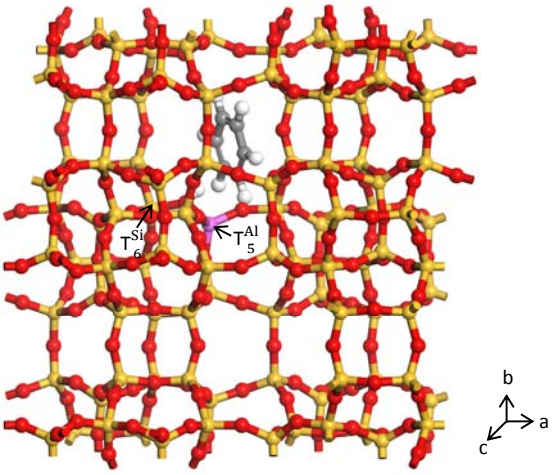
T^{Al}_{5}-O^{H}-T^{Si}_{1}



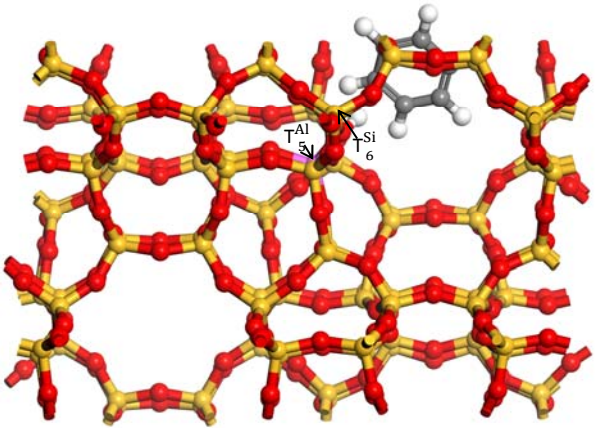


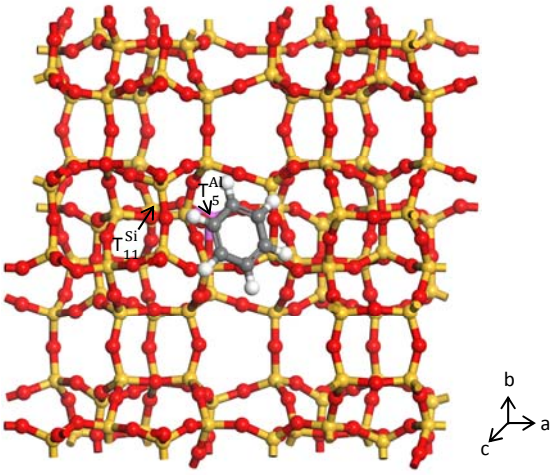
T^{Al}_{5}-O^{H}-T^{Si}_{4}



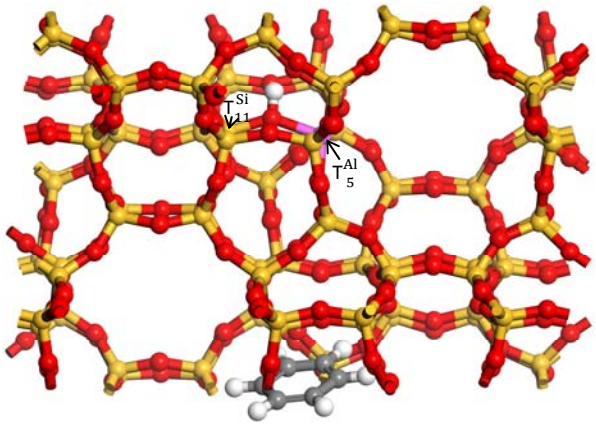


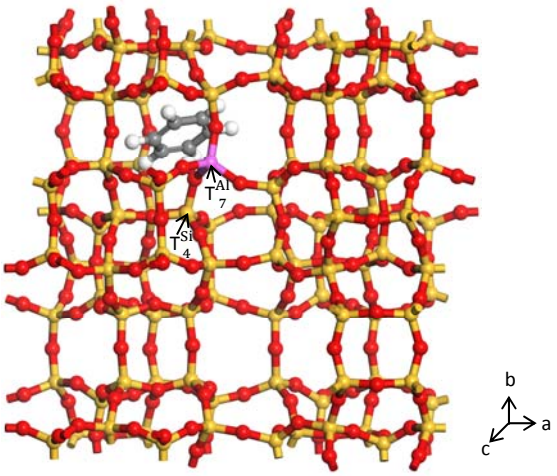
T^{Al}_{5}-O^{H}-T^{Si}_{6}



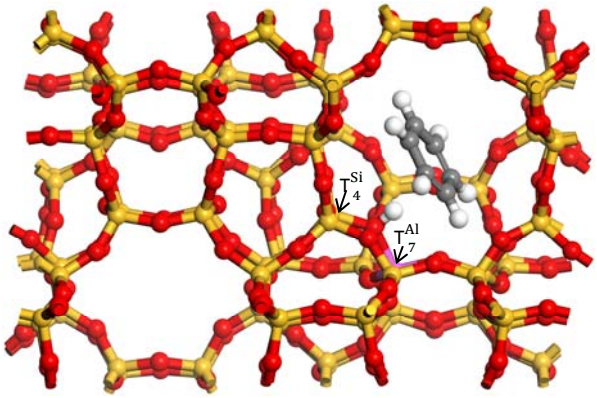


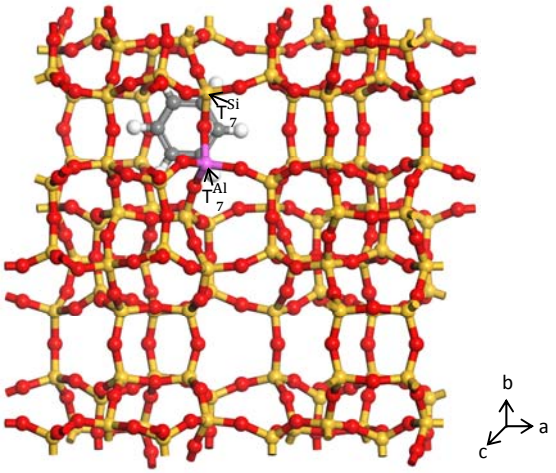
T^{Al}_{5}-O^{H}-T^{Si}_{11}



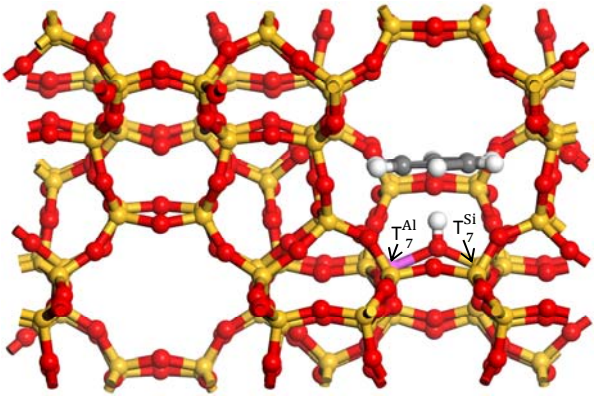


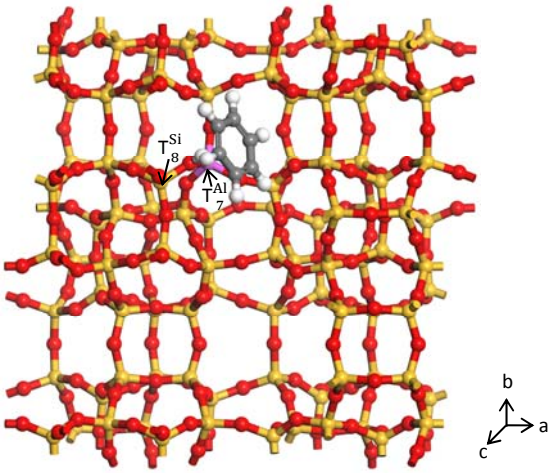
T_7^{Al}-O^H-T_4^{Si}



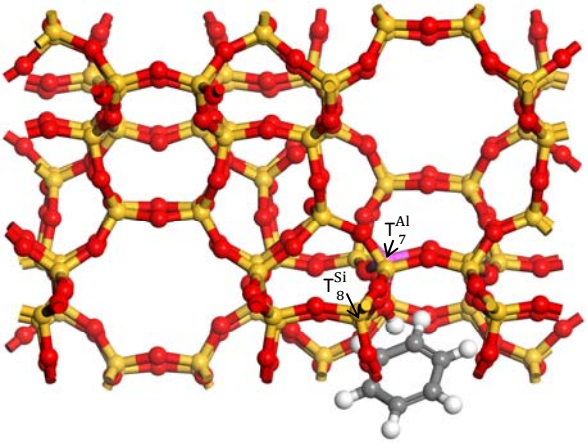


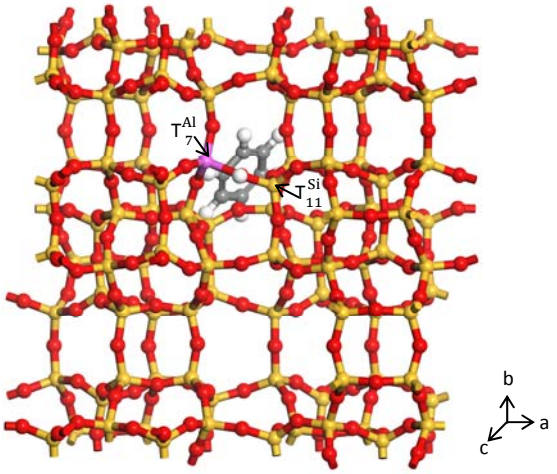
T_7^{Al}-O^H-T_7^{Si}



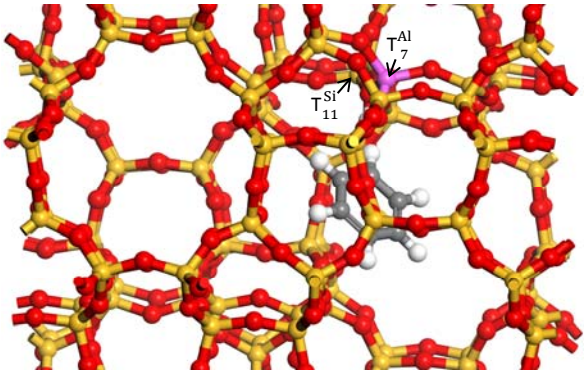


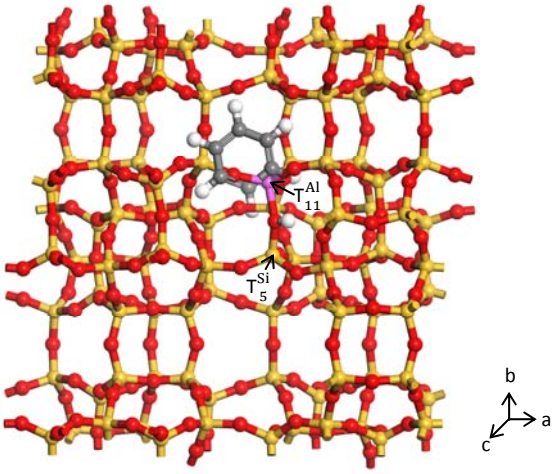
T_7^{Al}-O^H-T_8^{Si}



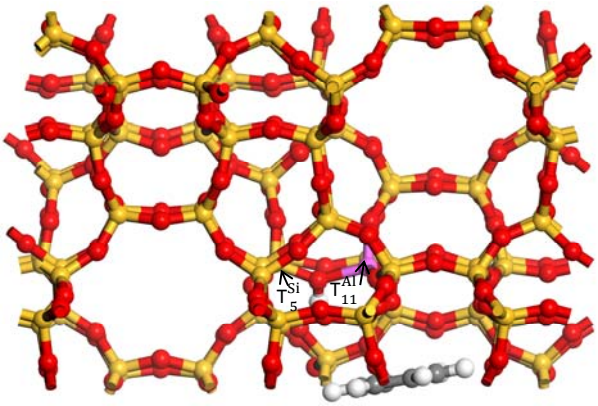


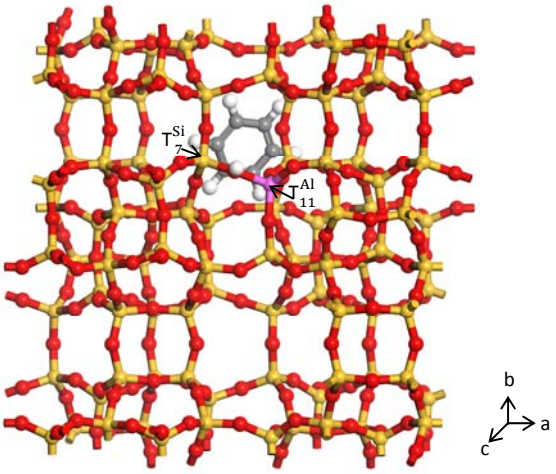
T_7^{Al}-O^H-T_{11}^{Si}



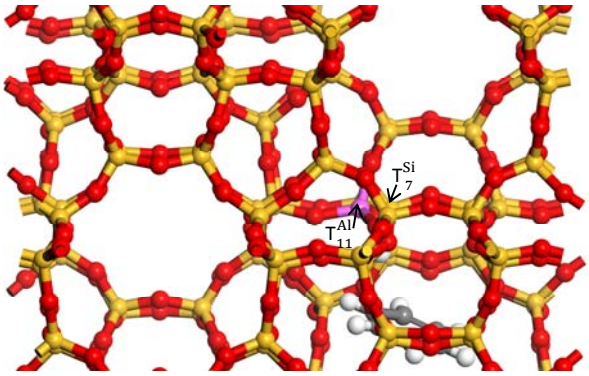


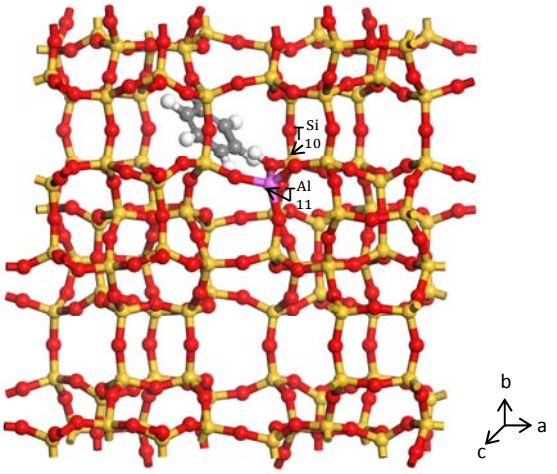
T_{11}^{Al}-O^{H}-T_{5}^{Si}



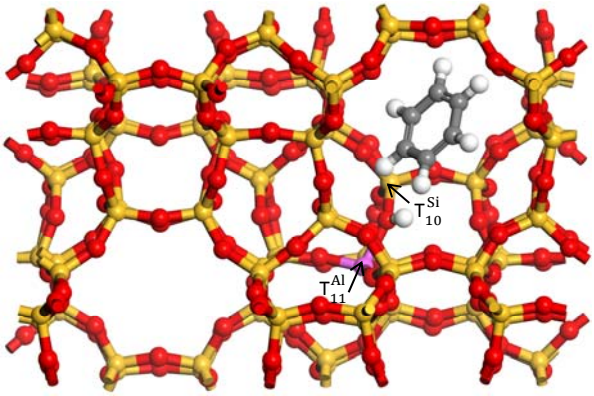


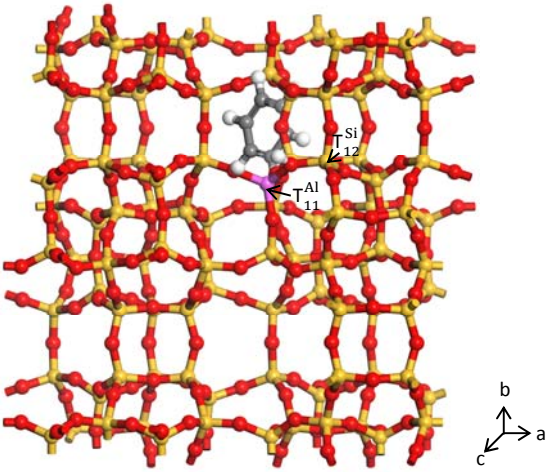
T_{11}^{Al}-O^{H}-T_{7}^{Si}



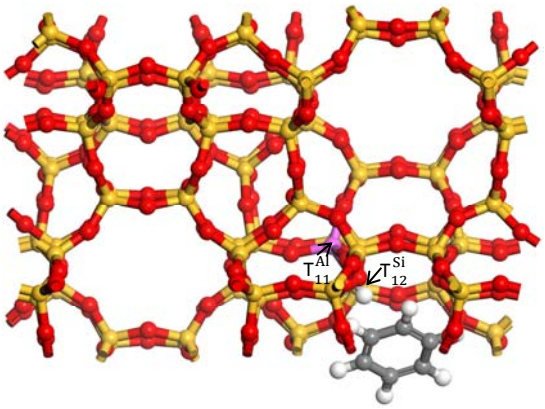


T_{11}^{Al}-O^{H}-T_{10}^{Si}





T_{11}^{Al}-O^{H}-T_{12}^{Si}



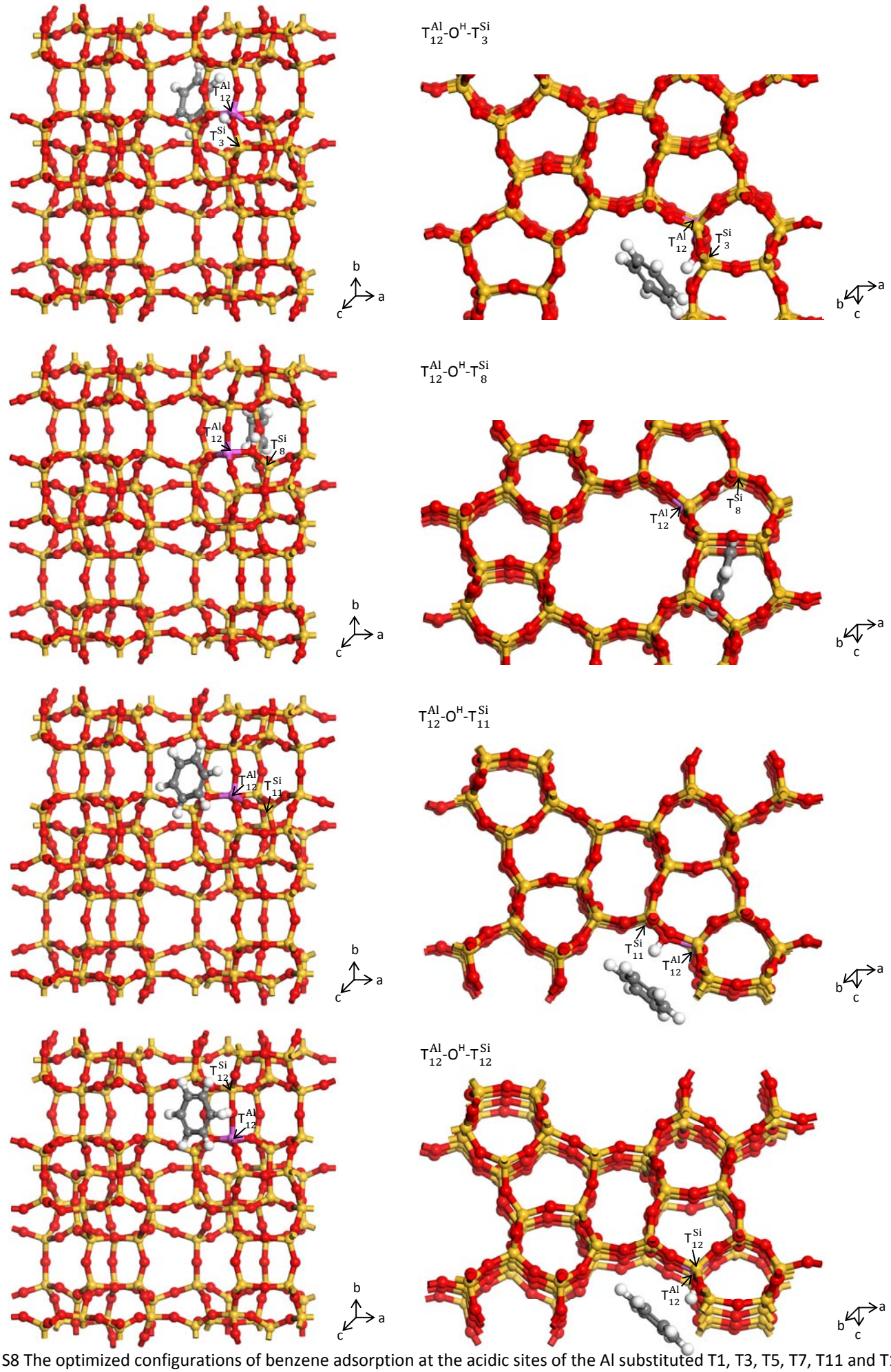
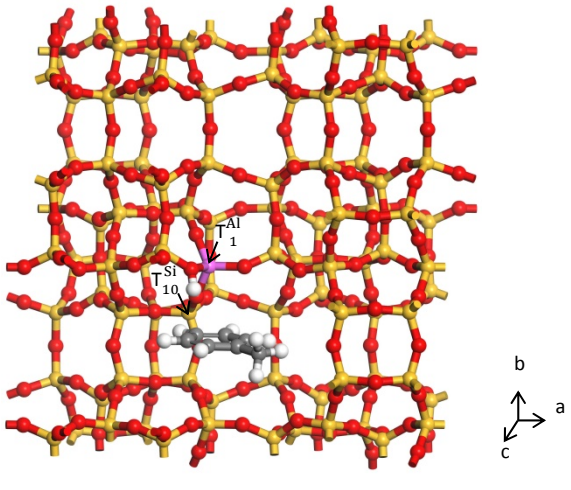
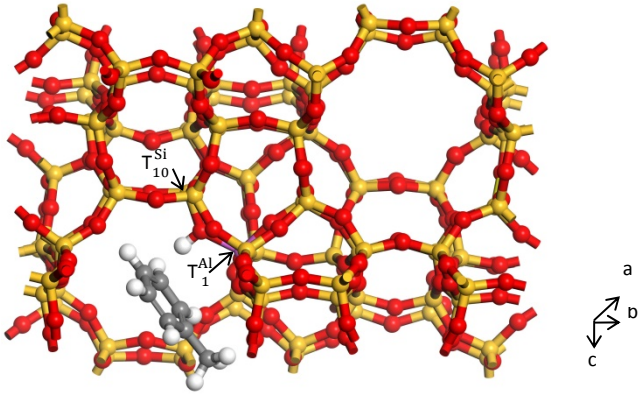
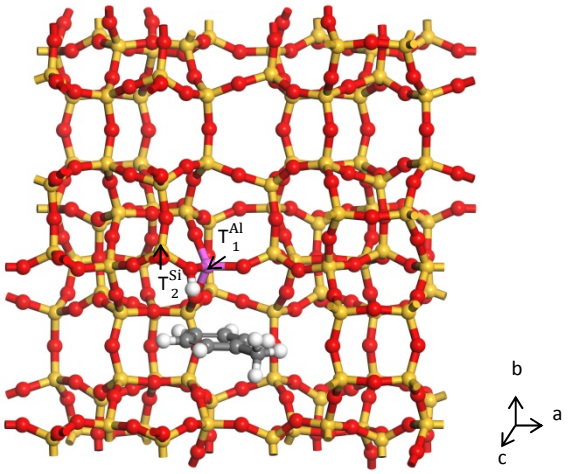


Figure S8 The optimized configurations of benzene adsorption at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework

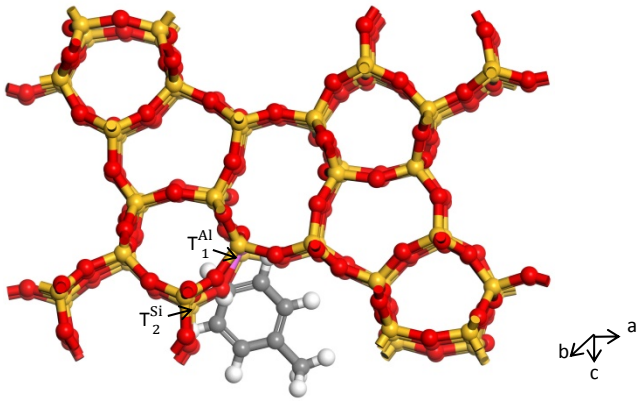


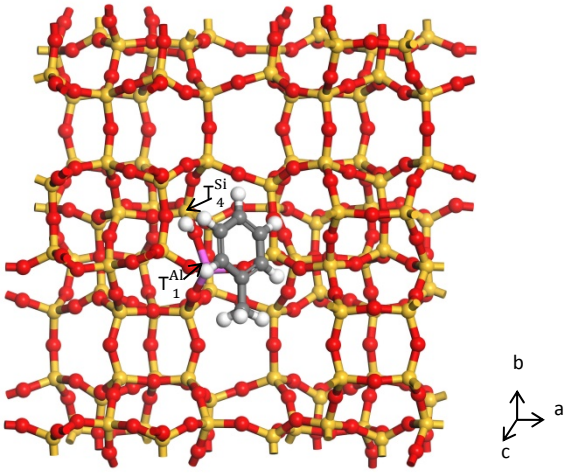
T^{Al}_{1}-O^{H}-T^{Si}_{10}



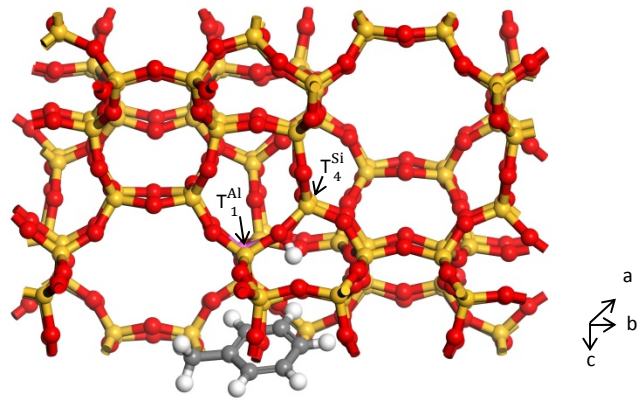


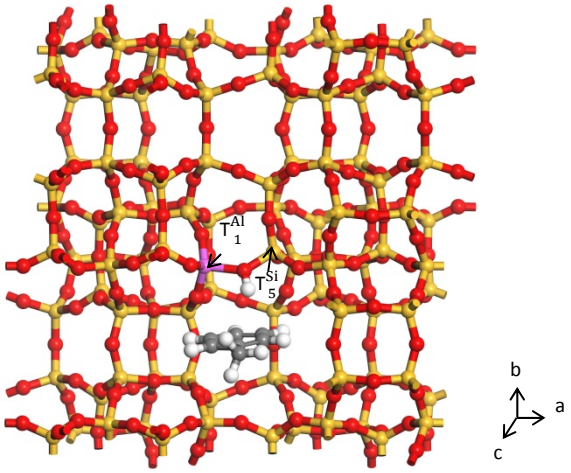
T^{Al}_{1}-O^{H}-T^{Si}_{2}



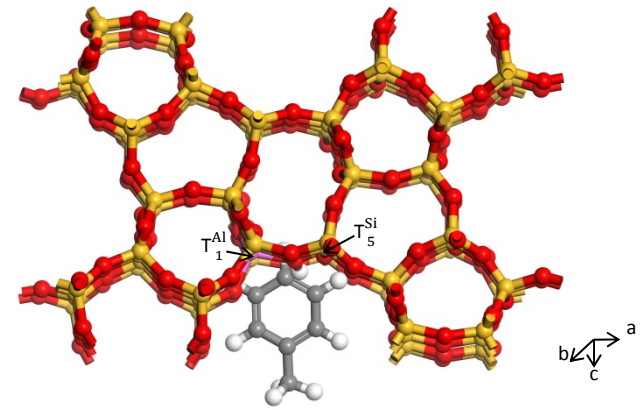


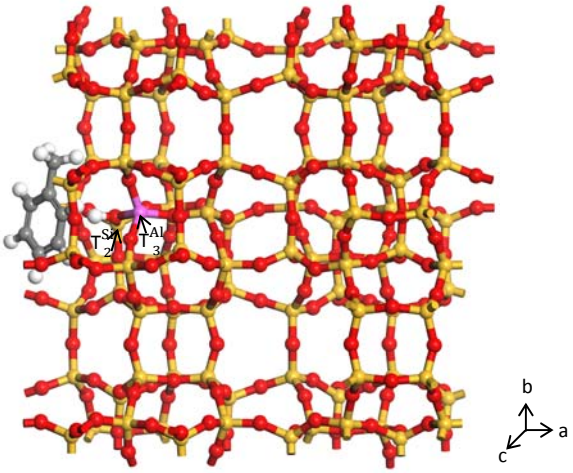
T^{Al}_{1}-O^{H}-T^{Si}_{4}



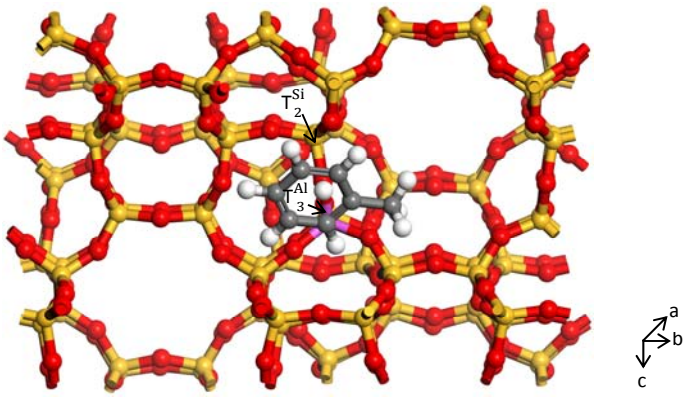


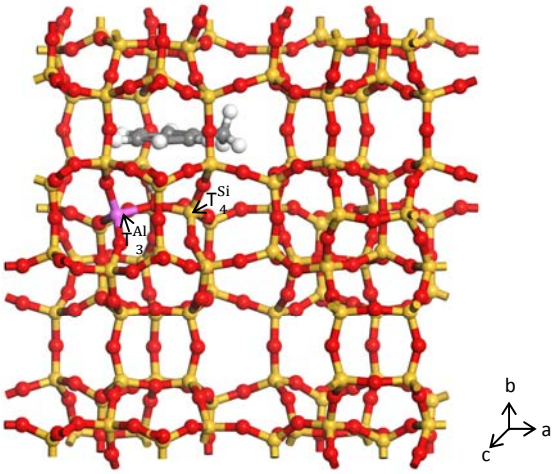
T^{Al}_{1}-O^{H}-T^{Si}_{5}



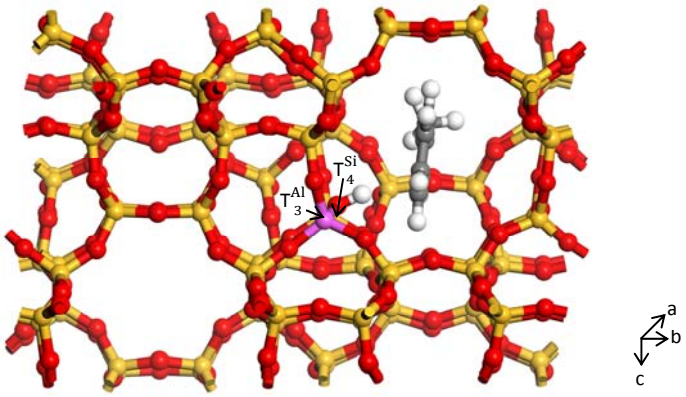


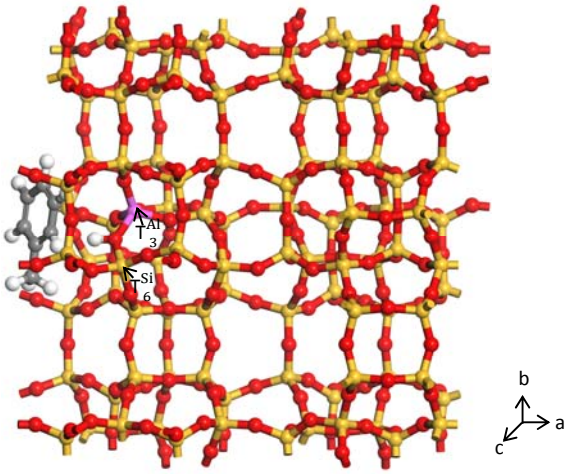
T^{Al}_{3}-O^{H}-T^{Si}_{2}



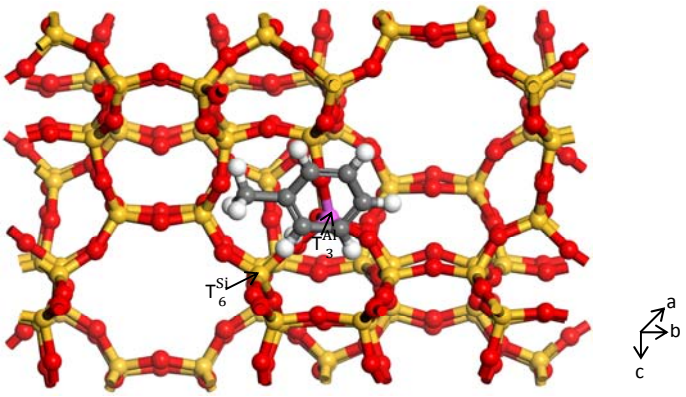


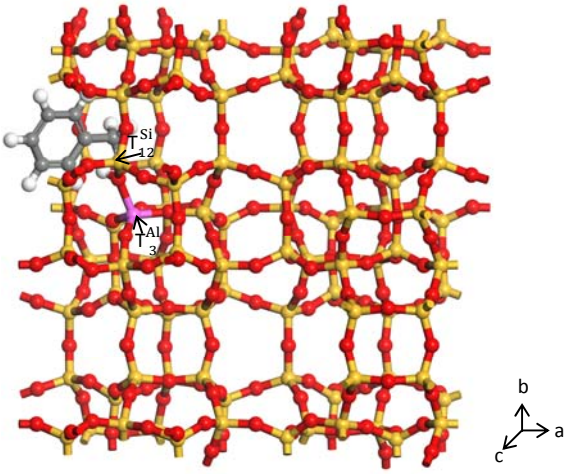
T^{Al}_{3}-O^{H}-T^{Si}_{4}



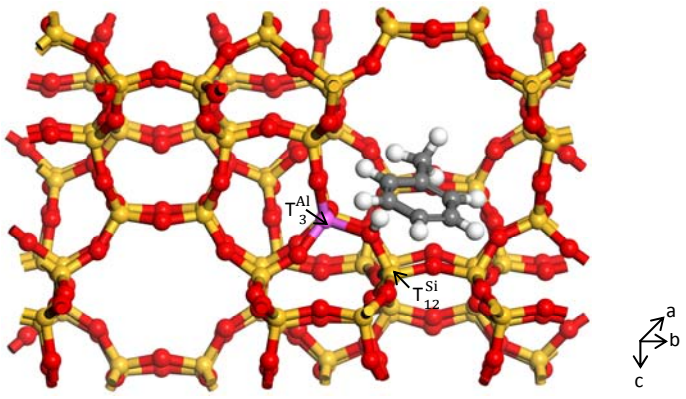


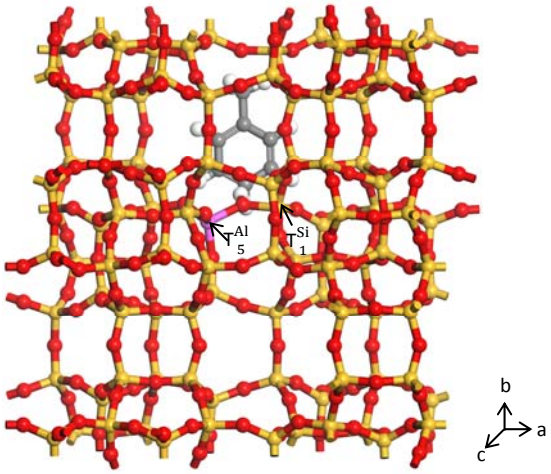
T^{Al}_{3}-O^{H}-T^{Si}_{6}



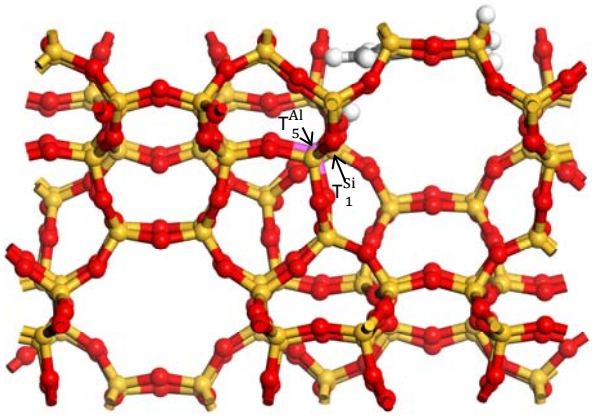


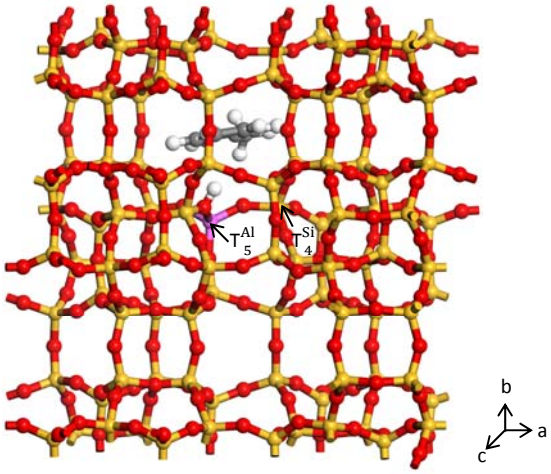
T^{Al}_{3}-O^{H}-T^{Si}_{12}



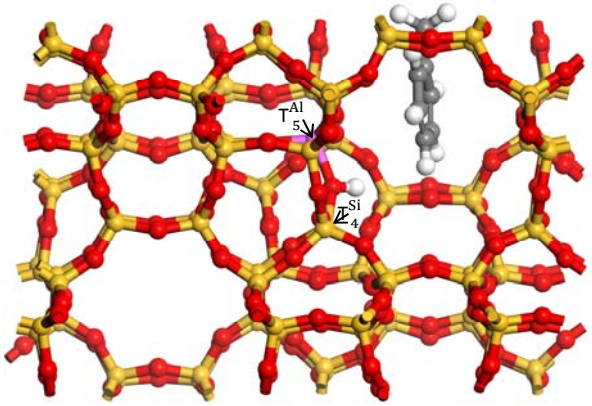


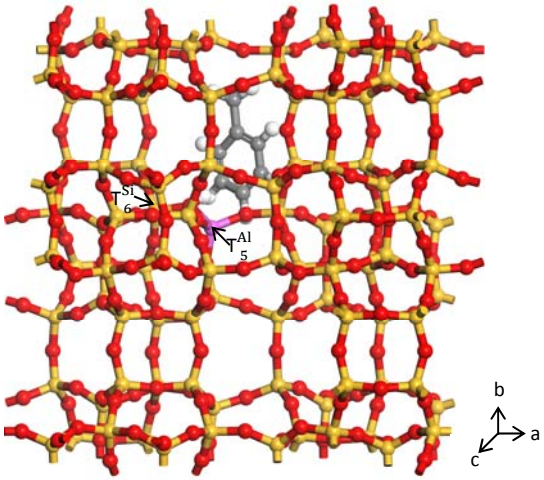
T^{Al}_{5}-O^{H}-T^{Si}_{1}



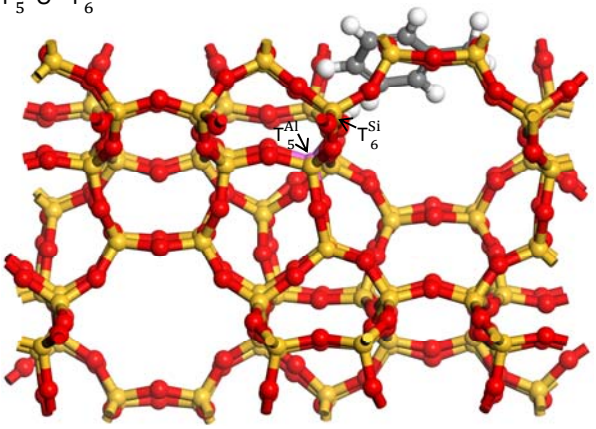


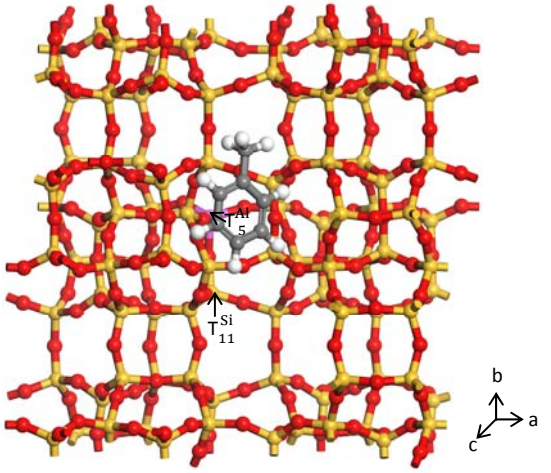
T^{Al}_{5}-O^{H}-T^{Si}_{4}



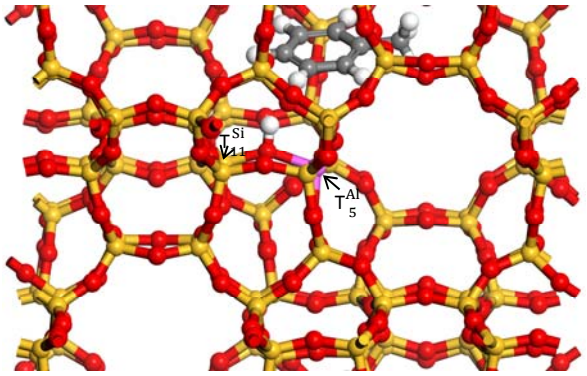


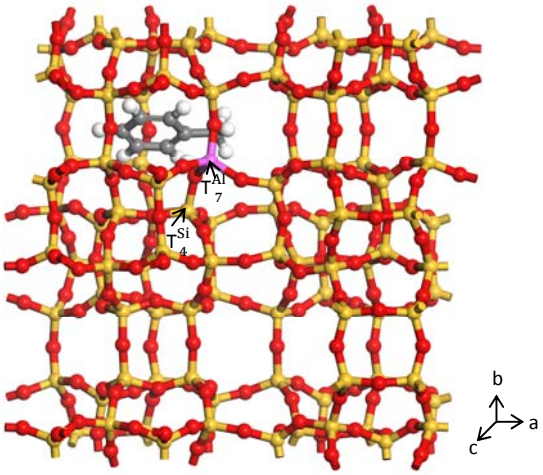
T^{Al}_{5}-O^{H}-T^{Si}_{6}



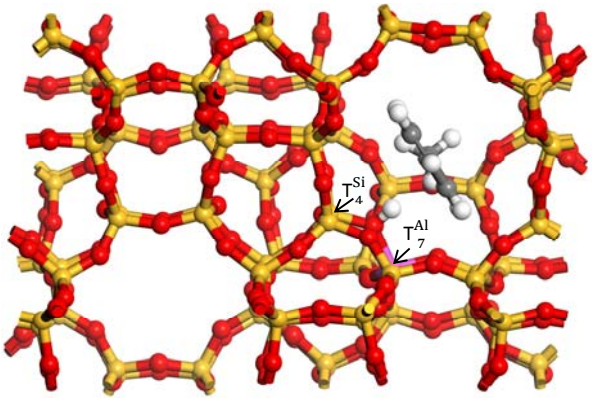


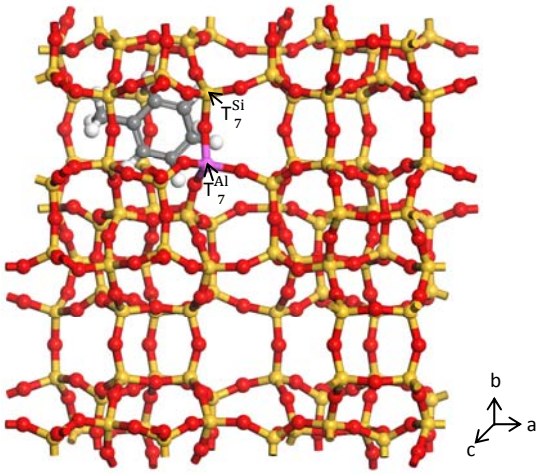
T^{Al}_{5}-O^{H}-T^{Si}_{11}



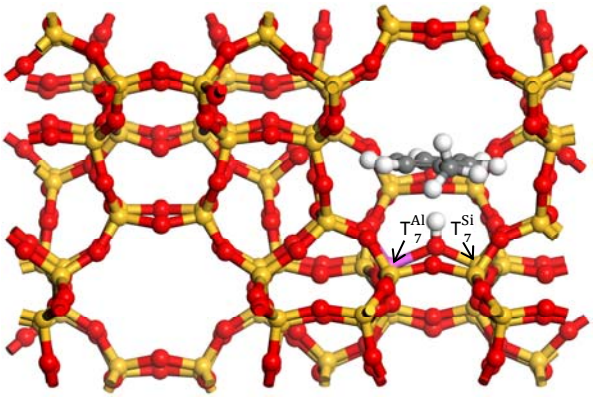


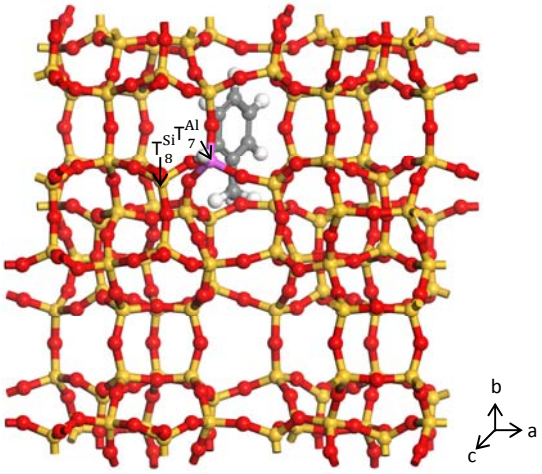
T^{Al}_{7}-O^{H}-T^{Si}_{4}



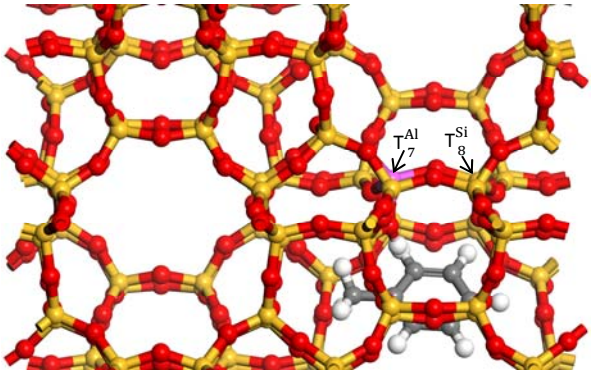


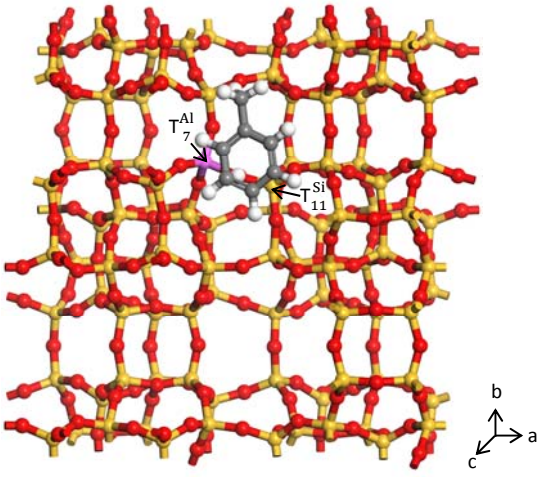
T^{Al}_{7}-O^{H}-T^{Si}_{7}



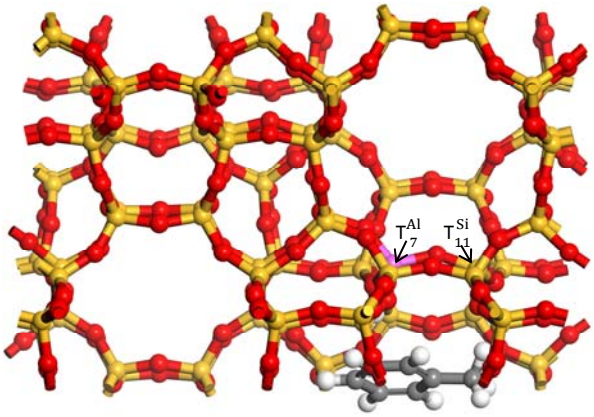


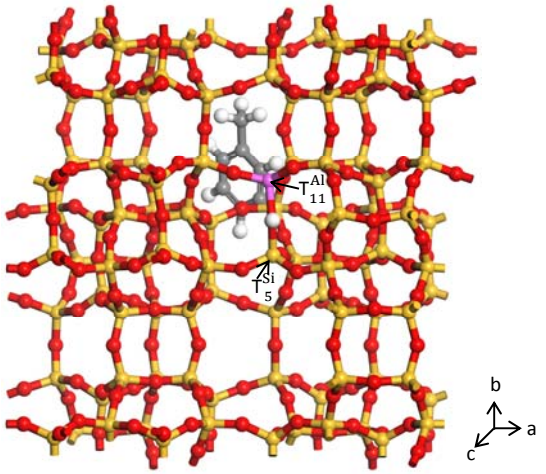
T^{Al}_{7}-O^{H}-T^{Si}_{8}



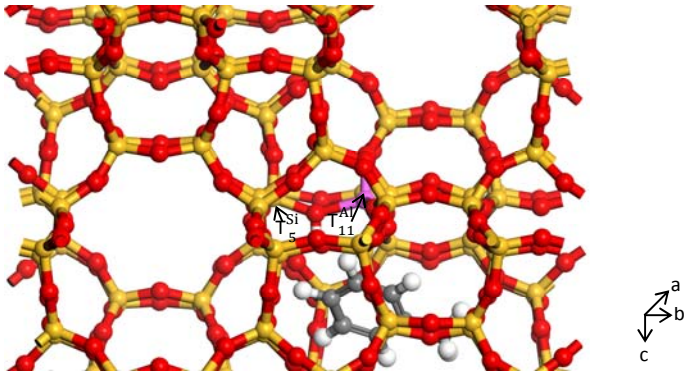


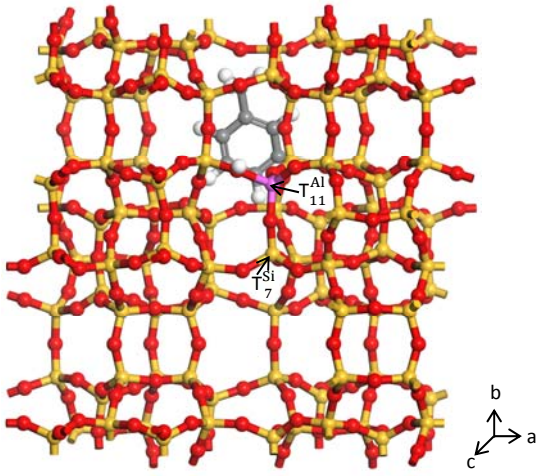
T^{Al}_{7}-O^{H}-T^{Si}_{11}



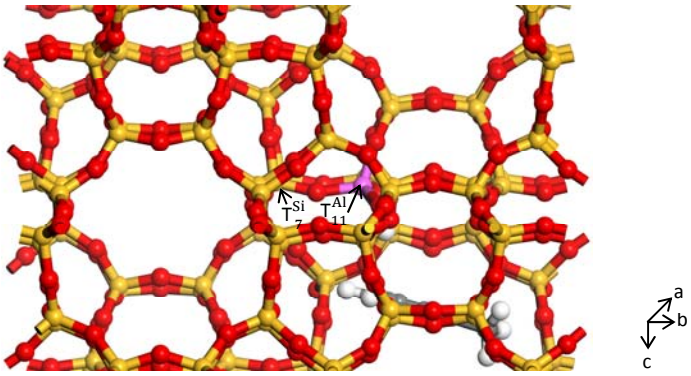


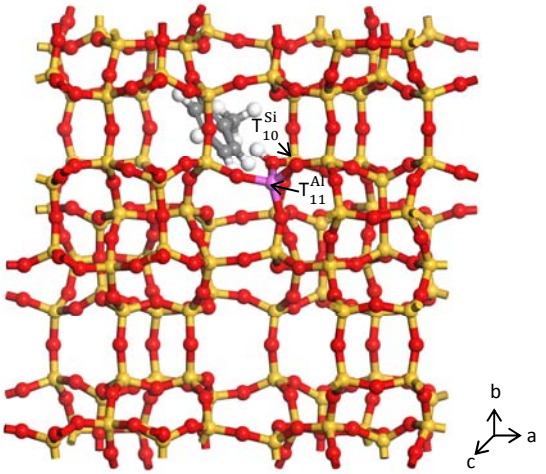
T^{Al}_{11}-O^{H}-T^{Si}_{5}



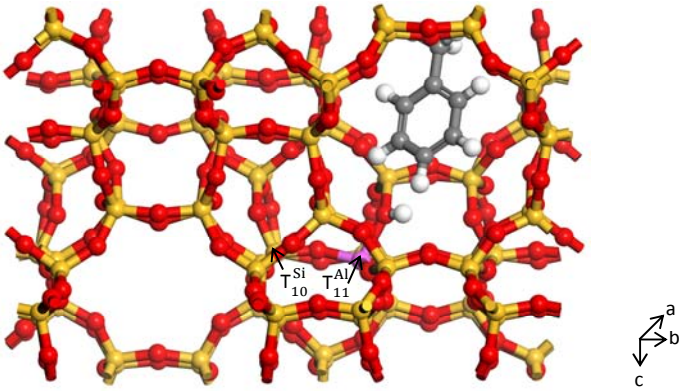


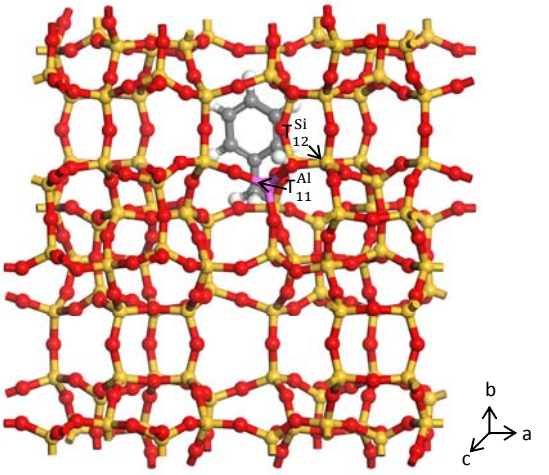
T^{Al}_{11}-O^{H}-T^{Si}_{7}



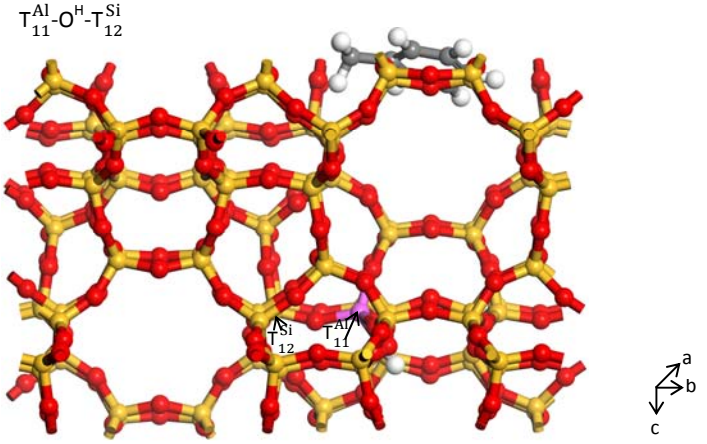


T^{Al}_{11}-O^{H}-T^{Si}_{10}





T^{Al}_{11}-O^{H}-T^{Si}_{12}



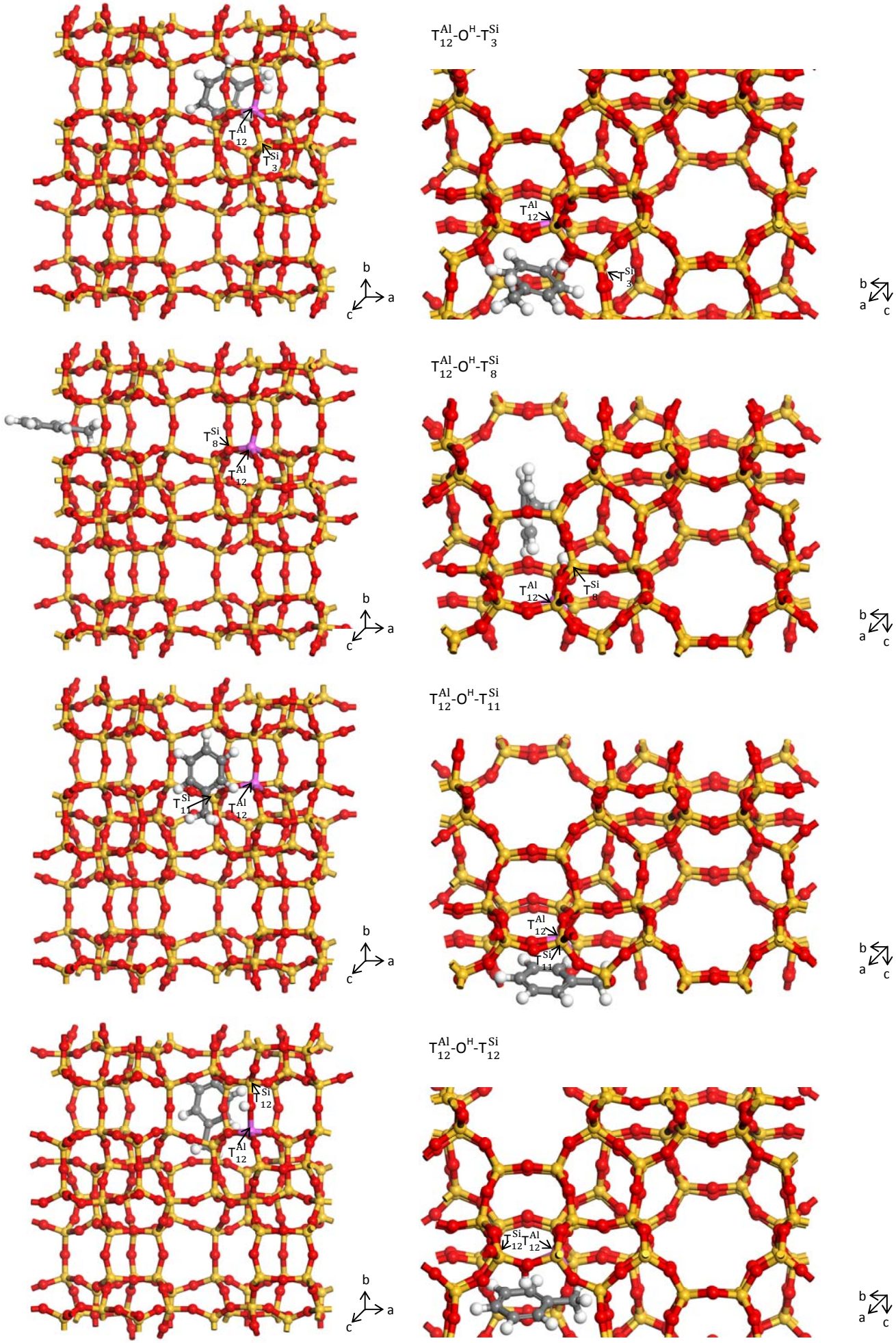
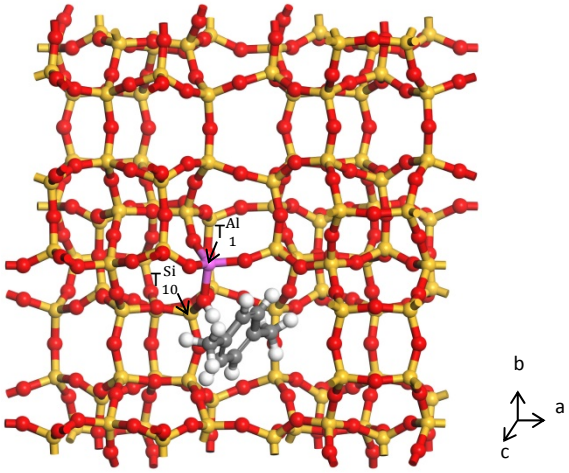
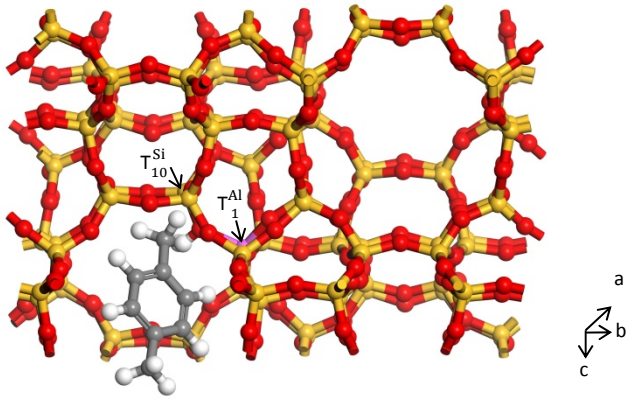
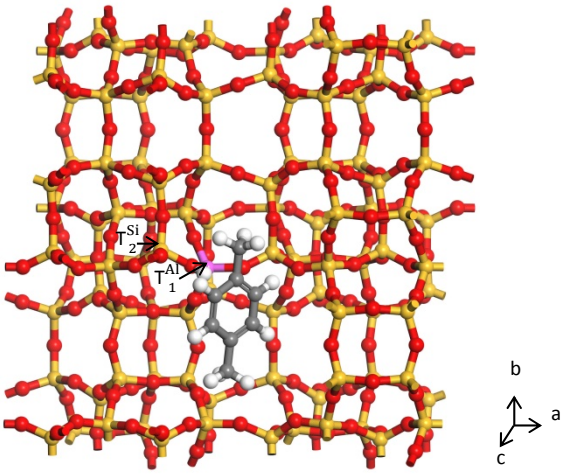


Figure S9 The optimized configurations of toluene adsorption at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework

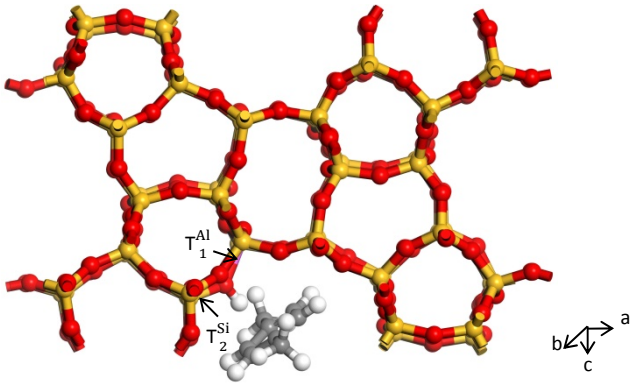


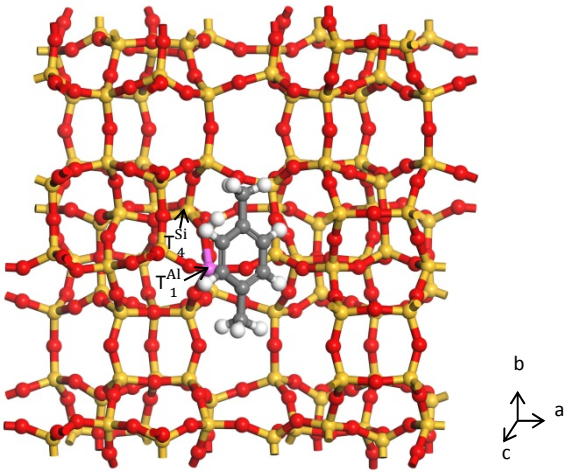
T^{Al}_{1}-O^{H}-T^{Si}_{10}



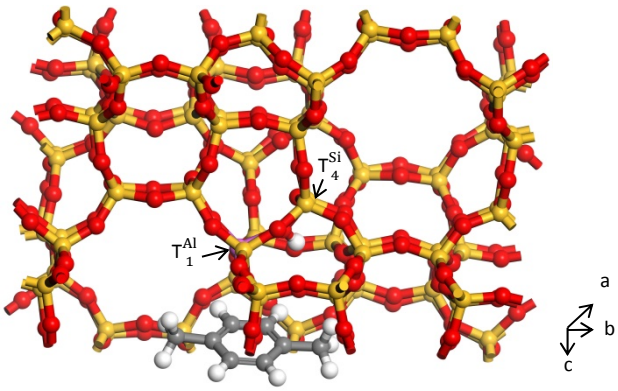


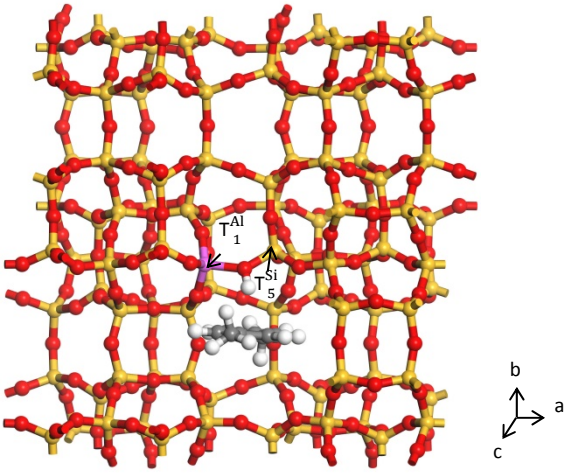
T^{Al}_{1}-O^{H}-T^{Si}_{2}



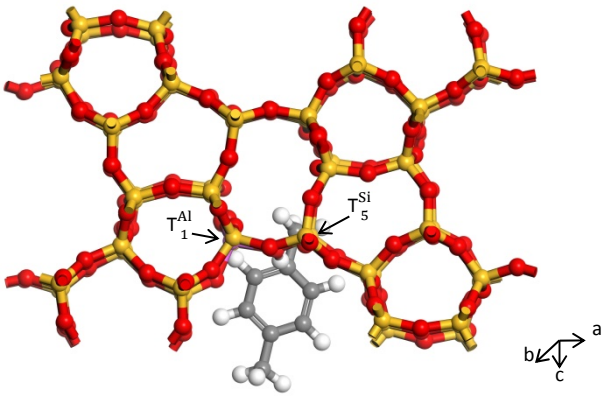


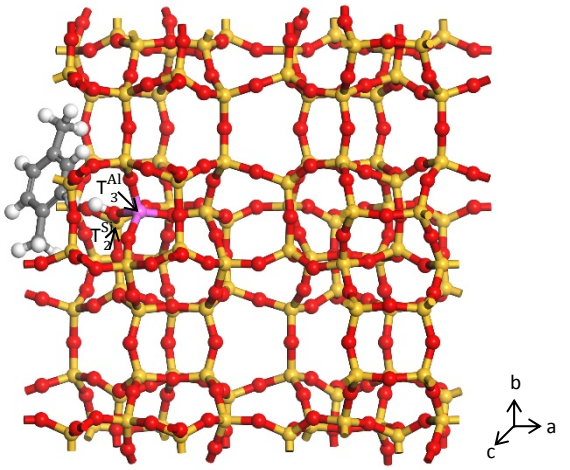
T^{Al}_{1}-O^{H}-T^{Si}_{4}



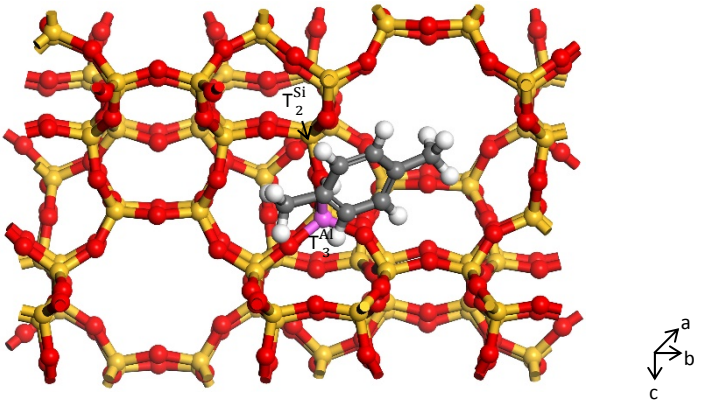


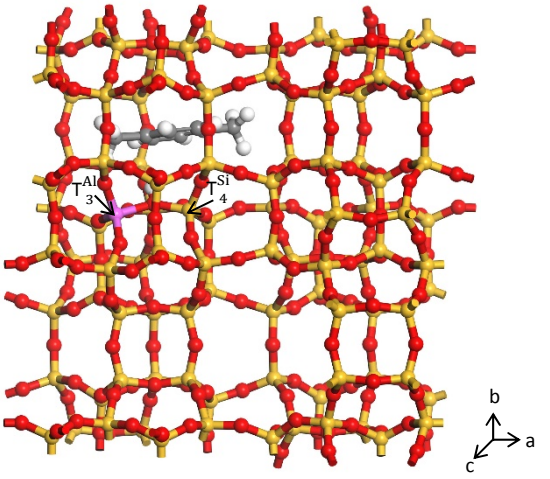
T^{Al}_{1}-O^{H}-T^{Si}_{5}



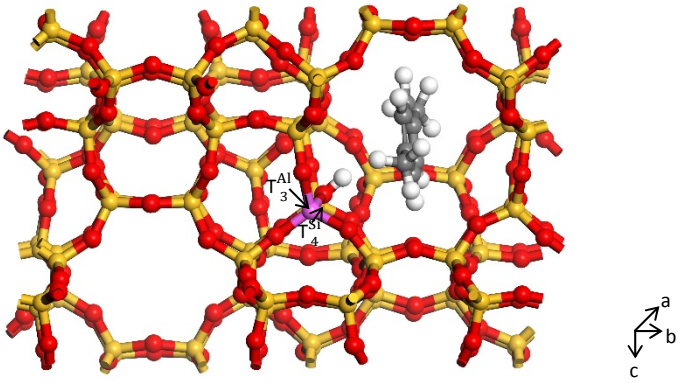


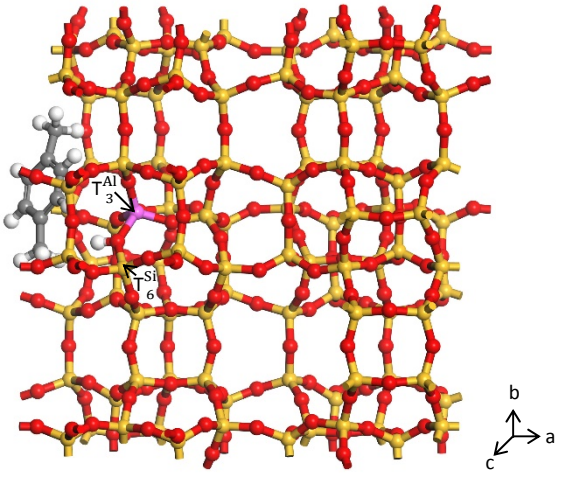
T^{Al}_{3}-O^{H}-T^{Si}_{2}



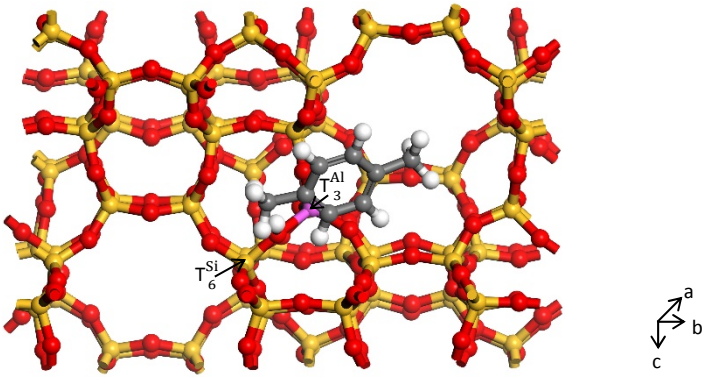


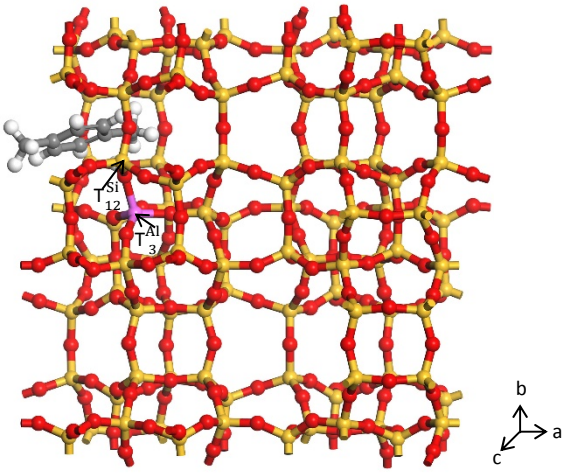
T^{Al}_{3}-O^{H}-T^{Si}_{4}



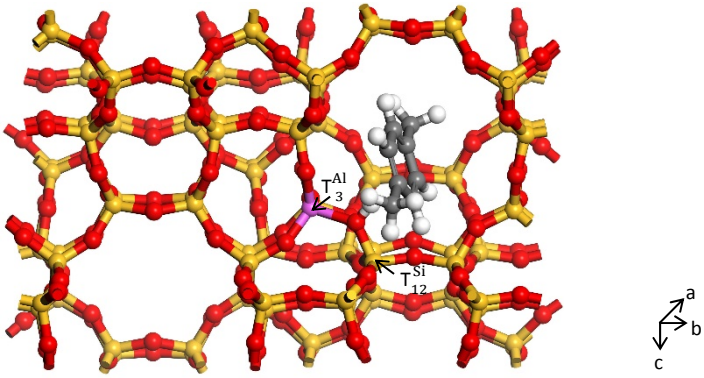


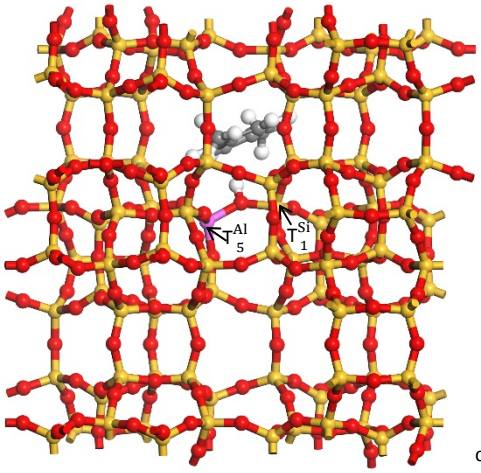
T^{Al}_{3}-O^{H}-T^{Si}_{6}



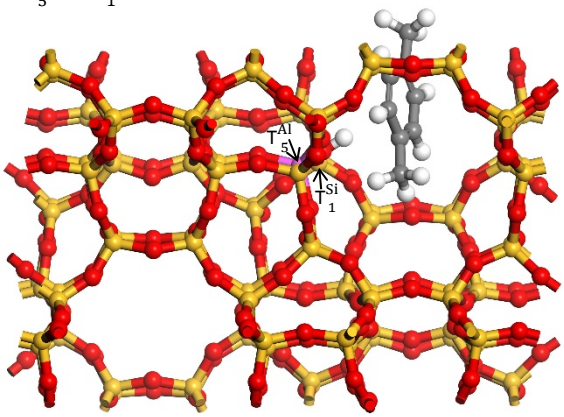


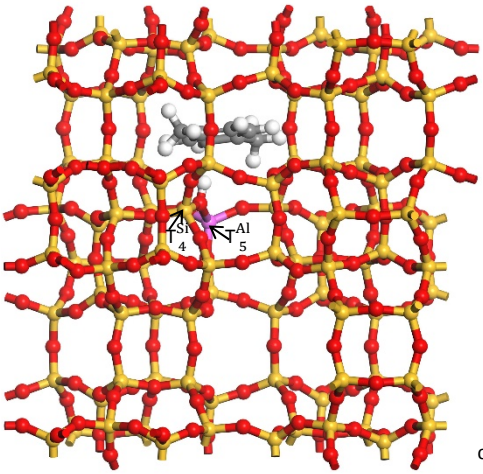
T^{Al}_{3}-O^{H}-T^{Si}_{12}



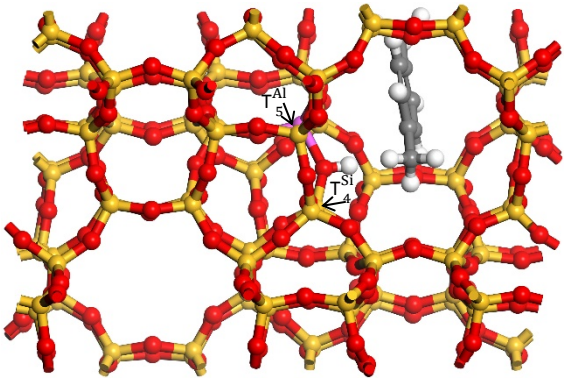


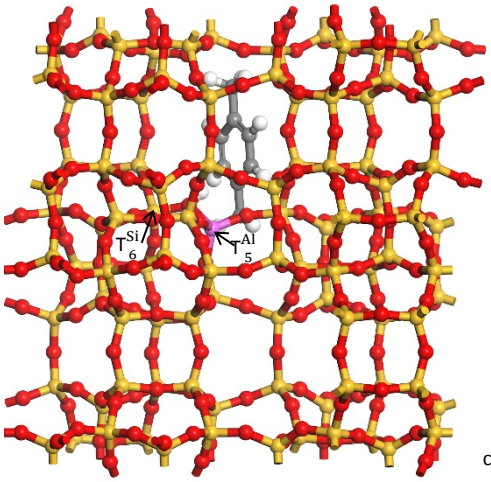
T^{Al}_{5}-O^{H}-T^{Si}_{1}



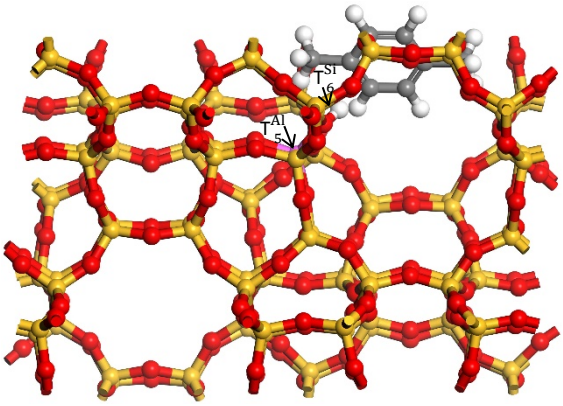


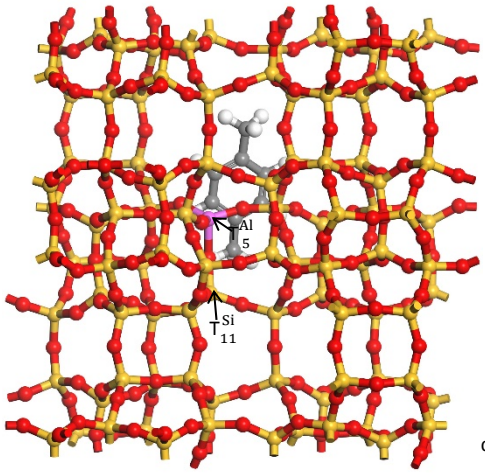
T^{Al}_{5}-O^{H}-T^{Si}_{4}



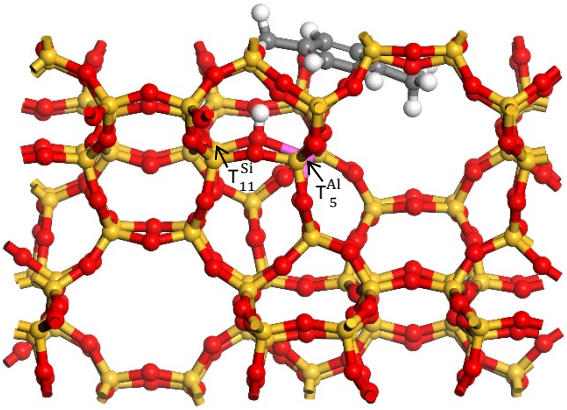


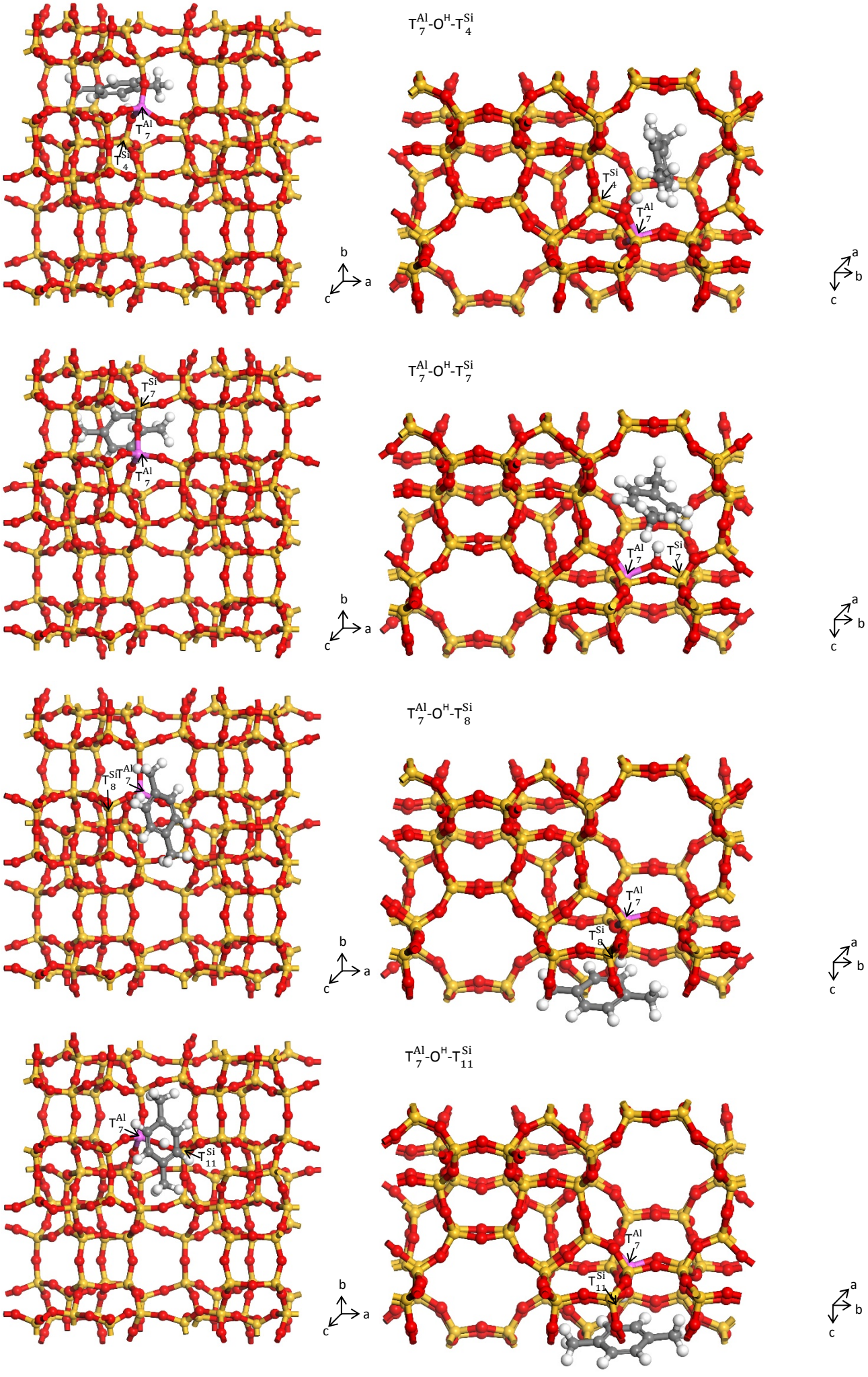
T^{Al}_{5}-O^{H}-T^{Si}_{6}

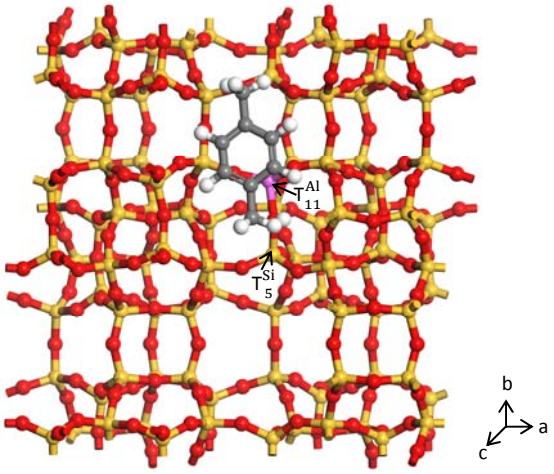




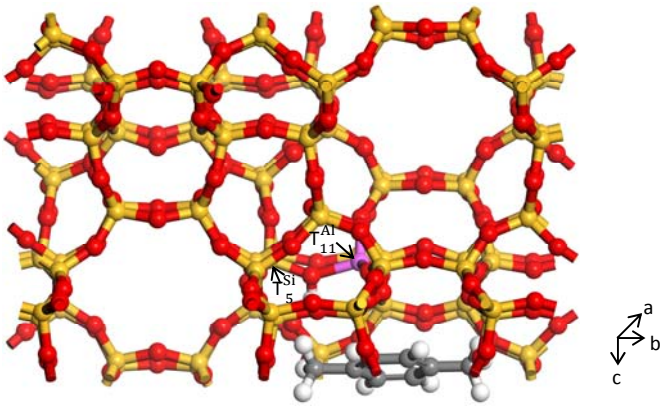
T^{Al}_{5}-O^{H}-T^{Si}_{11}

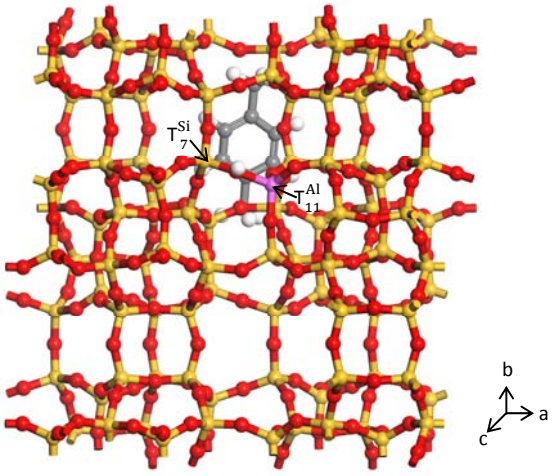



T_7^{Al}-O^H-T_4^{Si}
T_7^{Al}
T_4^{Si}
T_7^{Al}-O^H-T_7^{Si}
T_7^{Si}
T_7^{Al}
T_7^{Al}-O^H-T_8^{Si}
T_8^{Si}
T_7^{Al}
T_7^{Al}-O^H-T_{11}^{Si}
T_7^{Al}
T_{11}^{Si}
a
b
c

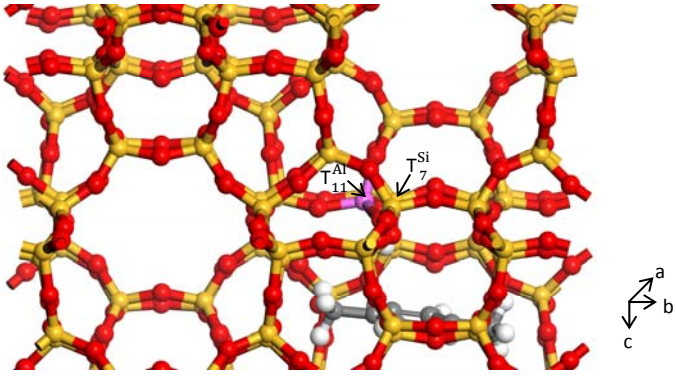


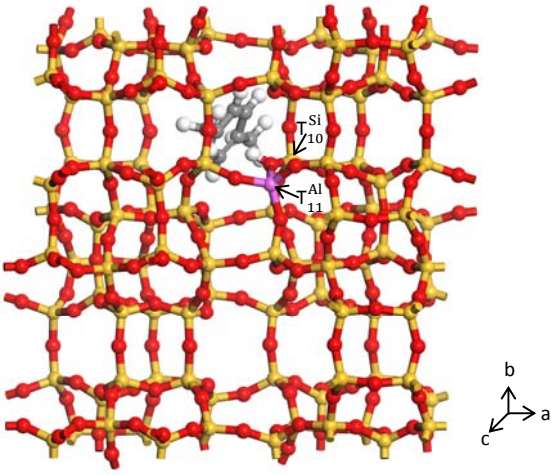
T_{11}^{Al}-O^{H}-T_{5}^{Si}



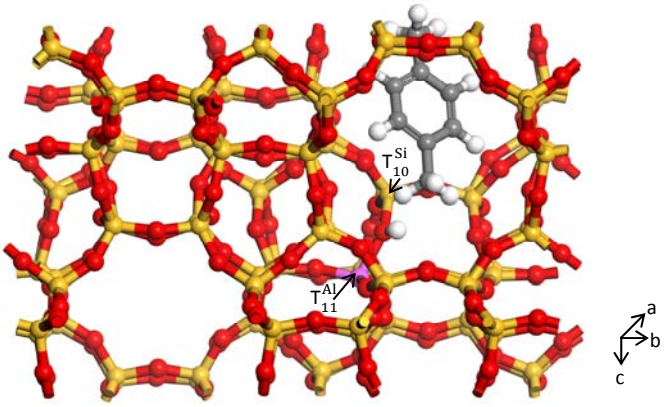


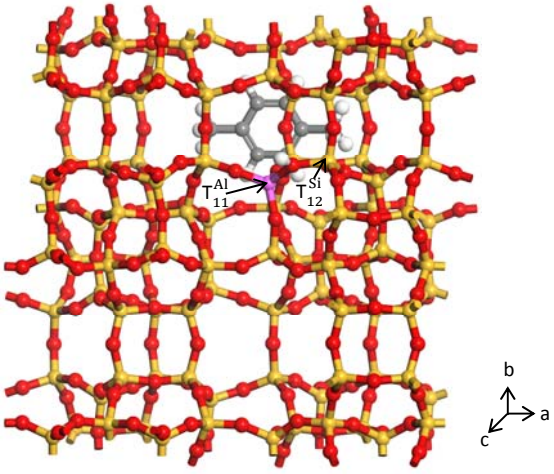
T_{11}^{Al}-O^{H}-T_{7}^{Si}



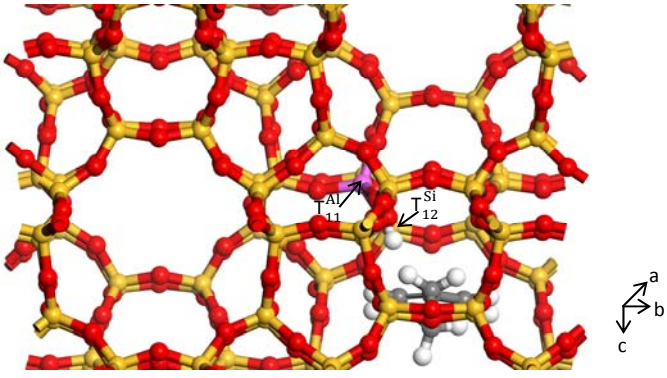


T_{11}^{Al}-O^{H}-T_{10}^{Si}





T_{11}^{Al}-O^{H}-T_{12}^{Si}



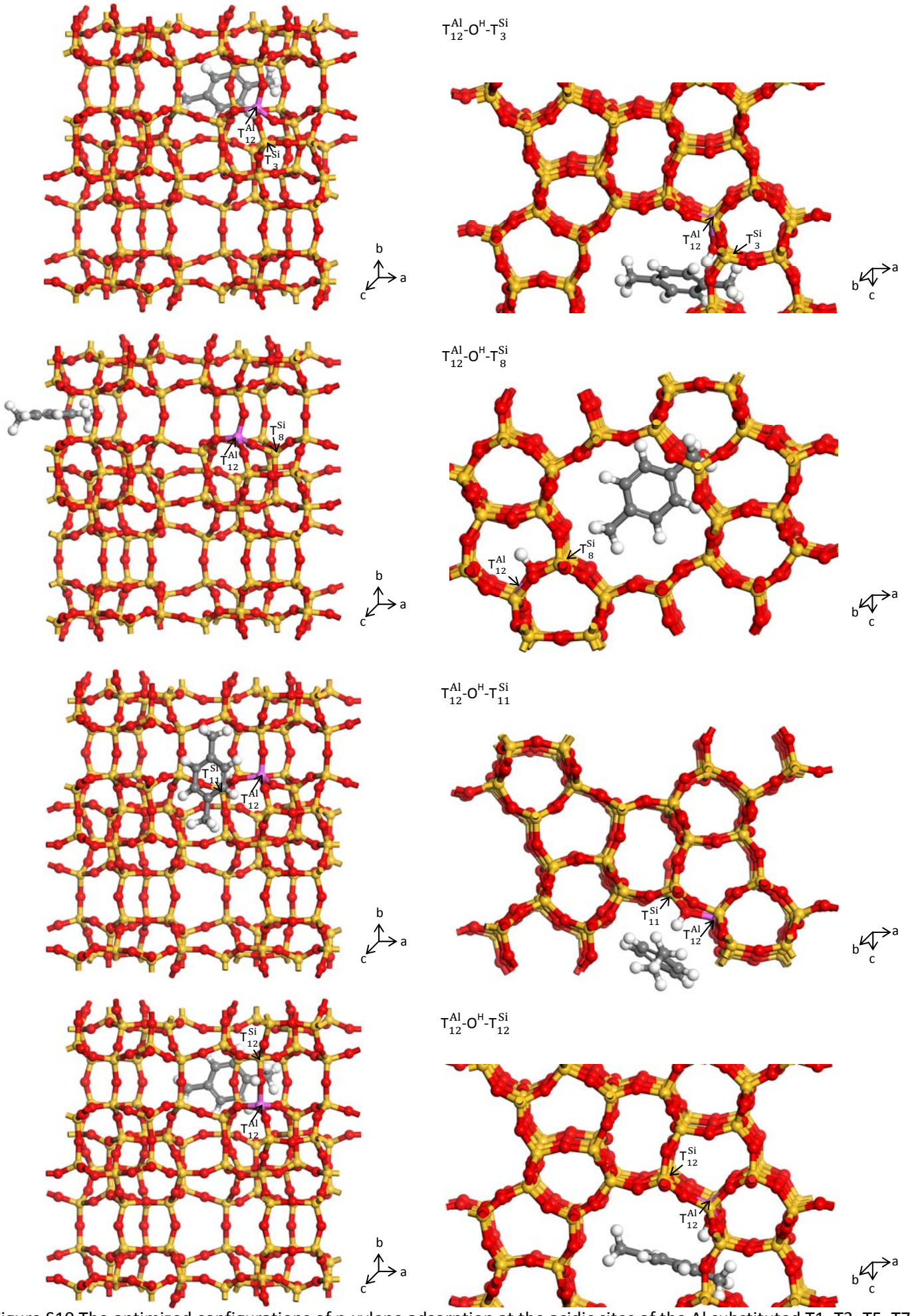


Figure S10 The optimized configurations of p-xylene adsorption at the acidic sites of the Al substituted T1, T3, T5, T7, T11 and T12 sites in the framework