Supporting Information:

The influence of adjacent Al atoms to the hydrothermal stability in H-SSZ-13: a first principles study

Lu Shi[a], Jiaqiang Yang[a], Gurong Shen[b], Yunkun Zhao[c], Rong Chen[d], Meiqing Shen[b], Yanwei Wen*[a] and Bin Shan*[a]

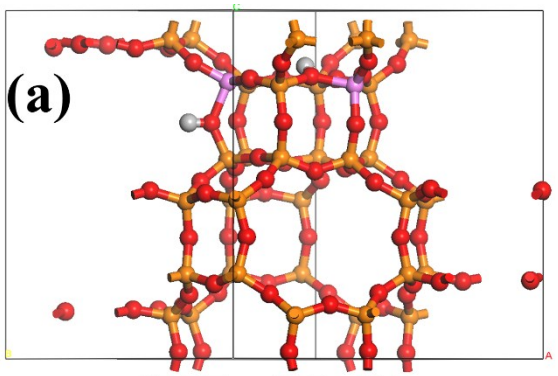
a. State Key Laboratory of Material Processing and Die and Mould Technology and School of Materials Science and Technology, Huazhong University of Science and Technology, Wuhan 430074, China.

E-mail: ywwen@mail.hust.edu.cn, bshan@mail.hust.edu.cn.

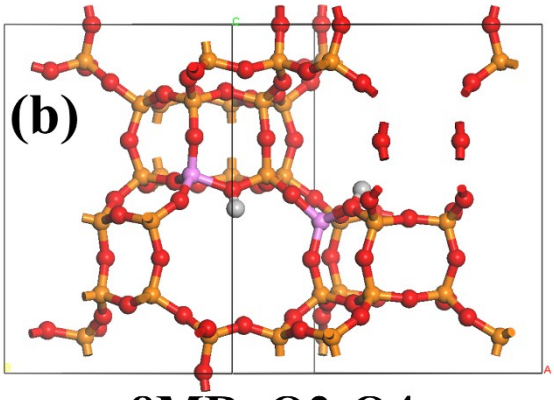
b. Key Laboratory for Green Chemical Technology of State Education Ministry, School of Chemical Engineering & Technology, Tianjin University, Tianjin 300072, China.

c. State Key Laboratory of Advanced Technologies for Comprehensive Utilization of Platinum Metal, Kunming Institute of Precious Metals, Kunming 650106, China.

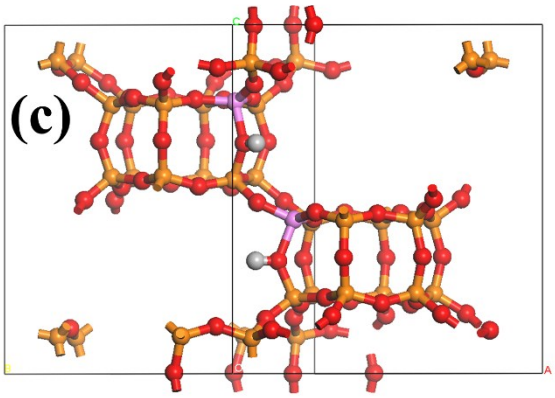
d. State Key Laboratory of Digital Manufacturing Equipment and Technology and School of Mechanical Science and Engineering, Huazhong University of Science and Technology, Wuhan 430074, China.



6MR_O2-O3



8MR_O2-O4



8MR_O1-O4

Figure S1. (a), (b) and (c) stand for the most stable Z_2H_2 configuration with 6MR_O2-O3, 8MR_O2-O4 and 8MR_O1-O4 frameworks, respectively.

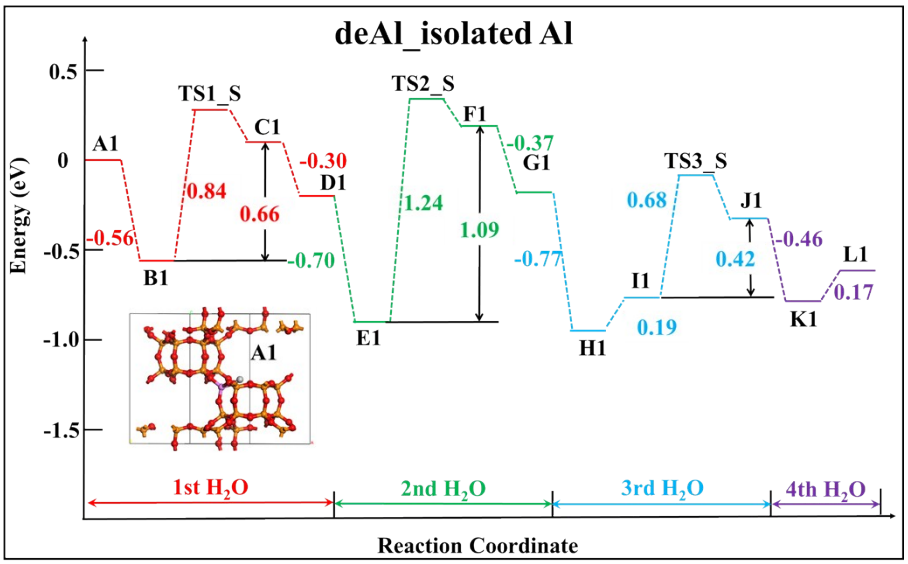


Figure S2. Detailed reaction energy and barriers of Al atom removed from the isolated Al framework in H-SSZ-13.

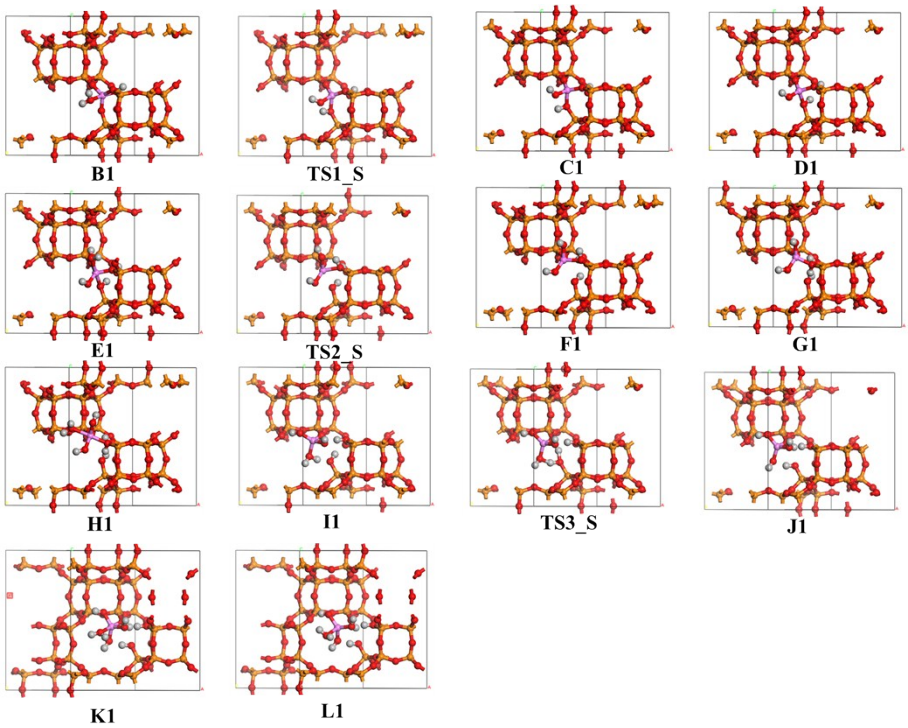


Figure S3. Reaction intermediates and transient structures for EFAL formation in the isolated Al framework, corresponding to Figure S2.

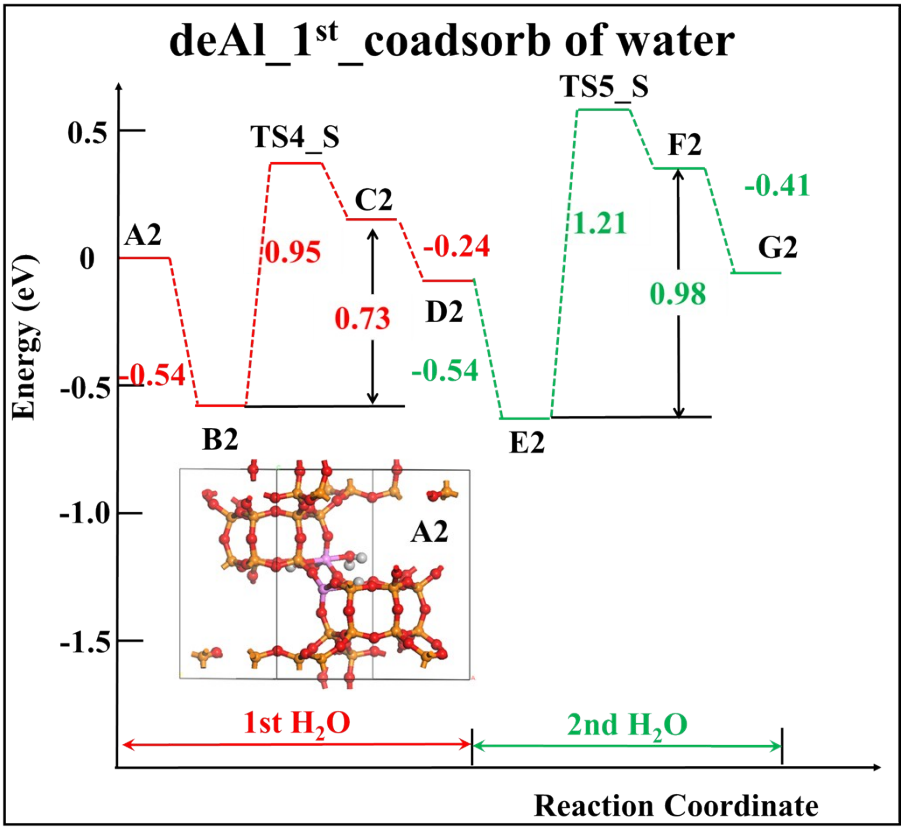


Figure S4. The reaction energy and barriers of the first two hydrolysis processes with two water molecules adsorb on different Al atoms.

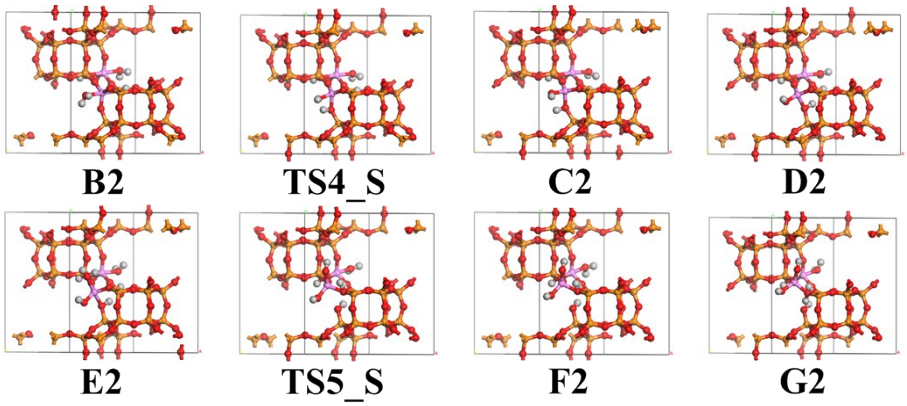


Figure S5. Reaction intermediates and transient structures for the first two hydrolysis processes with two water molecules adsorbed on different Al atoms, corresponding to Figure S4.

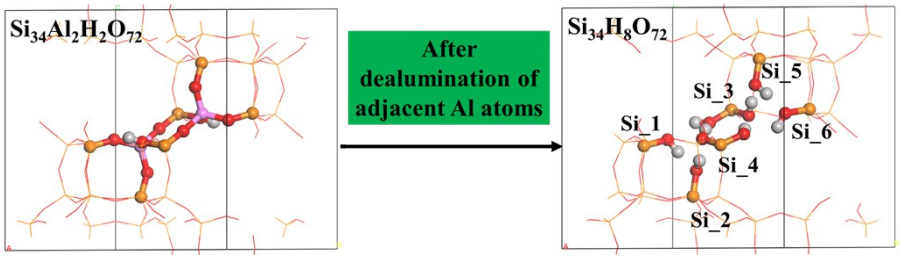


Figure S6. The transformation of adjacent Al atoms model ($Si_{34}Al_2H_2O_{72}$) to $Si_{34}H_8O_{72}$ after dealumination of adjacent Al atoms in the framework.

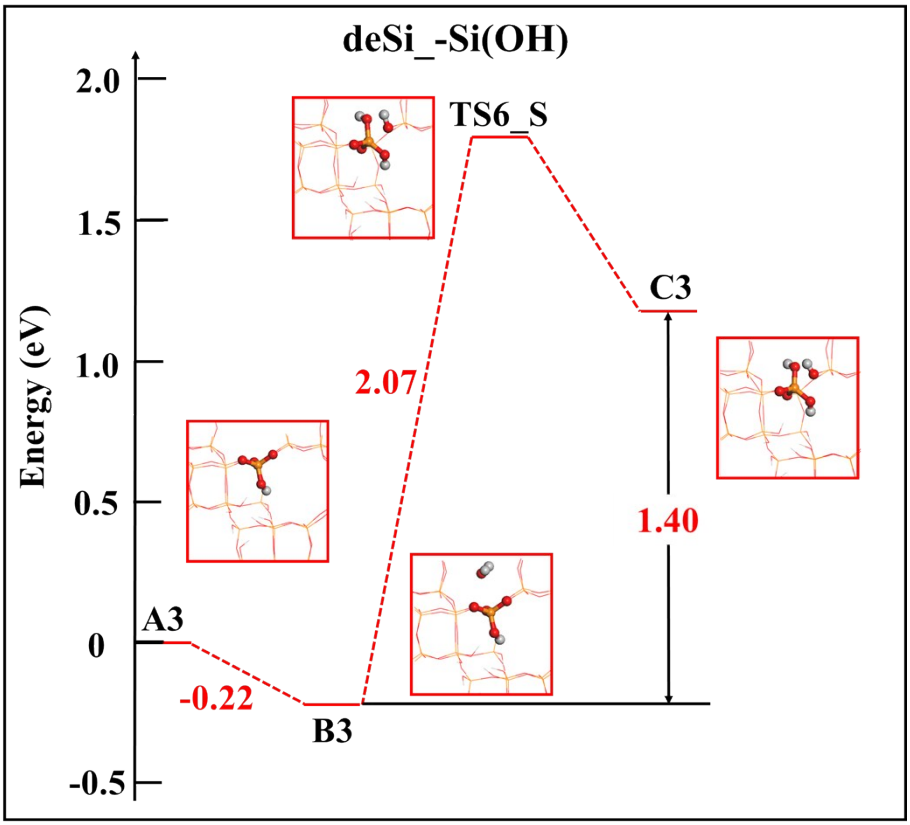


Figure S7. Detailed reaction energy and barrier of the first hydrolysis process during Si atom removed from the -Si(OH) local structure.

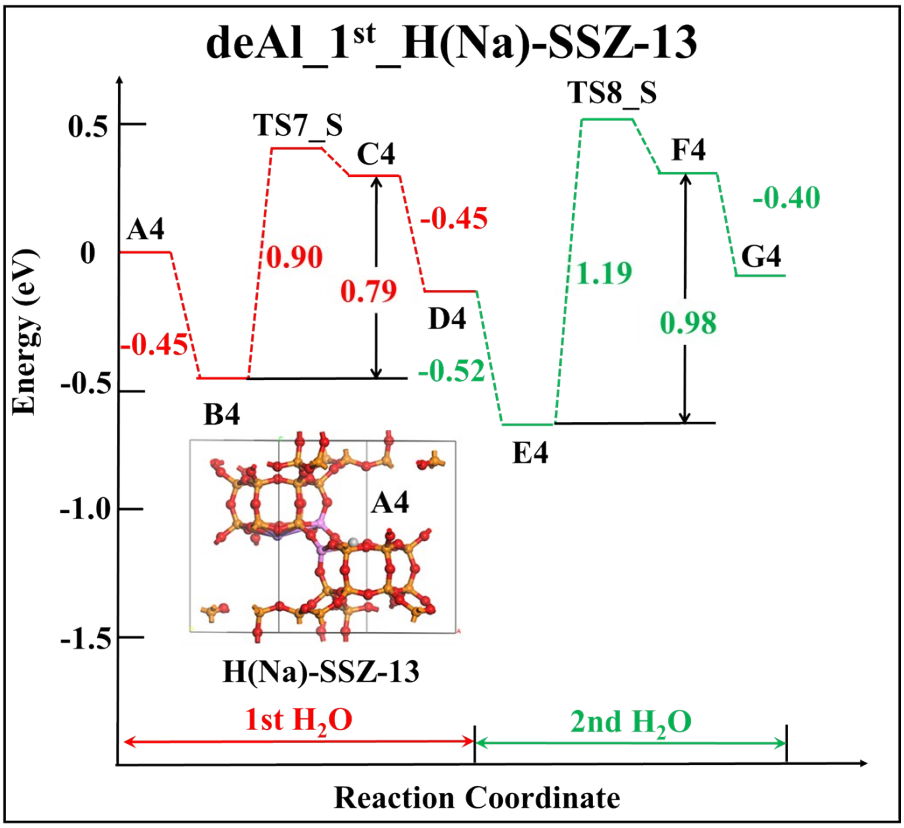


Figure S8. The reaction energy and barriers of the first two hydrolysis processes in H(Na)-SSZ-13.

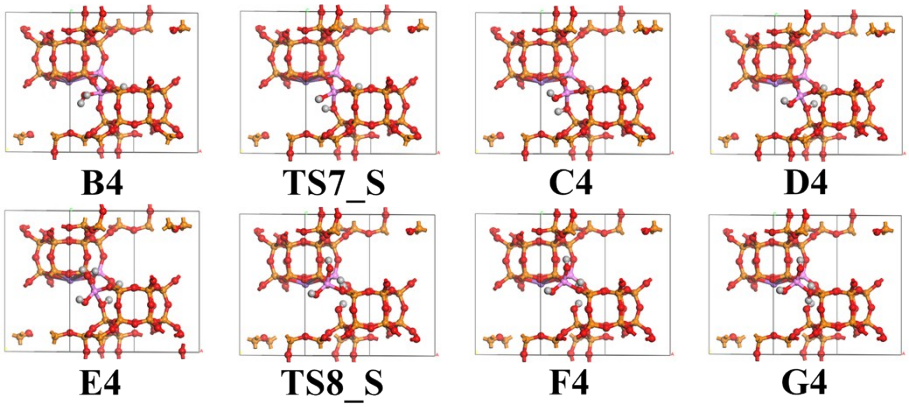


Figure S9. Reaction intermediates and transient structures for the first two hydrolysis processes in H(Na)-SSZ-13, corresponding to Figure S9.

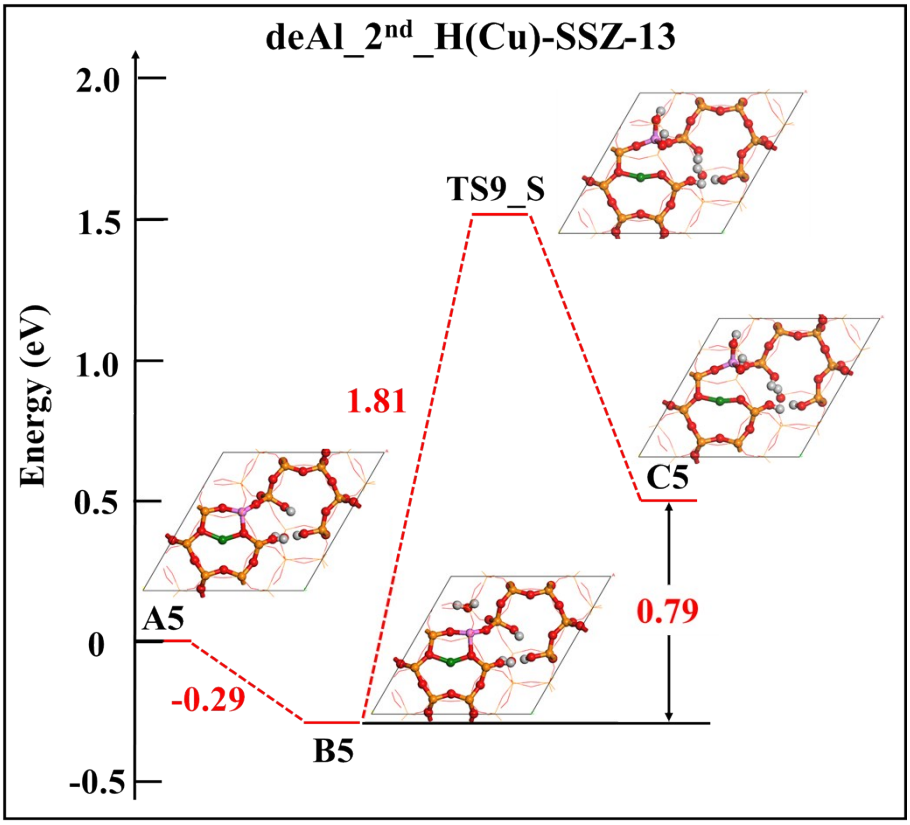


Figure S10. The detailed reaction barrier of first hydrolysis process with H(Cu)-SSZ-13, green ball = Cu.

Table S1. Number of relaxed atoms and imaginary frequencies calculated for each transition state.

Transition states	Number of relaxed atoms	Number of imaginary frequencies
TS1	9	1
TS2	10	1
TS3	11	1
TS4	9	1
TS5	8	1
TS6	11	1
TS7	10	2
TS8	11	2
TS9	9	1
TS1_S	9	1
TS2_S	8	1
TS3_S	11	1
TS4_S	9	1
TS5_S	10	1
TS6_S	9	2
TS7_S	9	1
TS8_S	10	1
TS9_S	9	1

INCAR files used for optimizations, transition states and frequencies calculation:

INCAR file used for optimizations
System = SSZ-13-dealumination
ISMEAR = 0
ISIF = 0
IVDW = 1
ENCUT = 400
ISPIN = 2
ALGO = FAST
NSW = 200
NELM = 99
IBRION = 2
LMAXMIX = 6
LWAVE = .FALSE.
LCHARG = .FALSE.
LPLANE = .FALSE.
EDIFF = 1E-6
EDIFFG = -0.02

INCAR file used for transition states search
System = TS
ISMEAR = 0
SIGMA = 0.1
ISIF = 0
IVDW = 1
ENCUT = 400
ISPIN = 2
ALGO = FAST
NSW = 1000
NELM = 99
IBRION = 3
LWAVE = .FALSE.
LCHARG = .FALSE.
LPLANE = .FALSE.
EDIFF = 1E-6
EDIFFG = -0.02
NPAR = 2
POTIM = 0.05
ICHAIN = 0
IMAGES = 6
SPRING = -5
LCLIMB = .T.

INCAR file used for frequency calculation
System = freq
ISMEAR = 0
ISIF = 0
IVDW = 1
ENCUT = 400
ISPIN = 2
ALGO = FAST
NSW = 1
NELM = 99
IBRION = 5
LMAXMIX = 6
LWAVE = .FALSE.
LCHARG = .FALSE.
LPLANE = .FALSE.
EDIFF = 1E-6
EDIFFG = -0.02
POTIM = 0.01
NFREE = 2