

Supporting Information

Mechanistic understanding of dehydroxylation reaction of smectites: Insights from reactive force field (ReaxFF) molecular dynamics simulation

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This file contains 11 pages, including 9 figures.

Contents

Figure S1.....S3

Figure S2.....S4

Figure S3.....S5

Figure S4.....S6

Figure S5.....S7

Figure S6.....S8

Figure S7.....S9

Figure S8.....S10

Figure S9.....S11

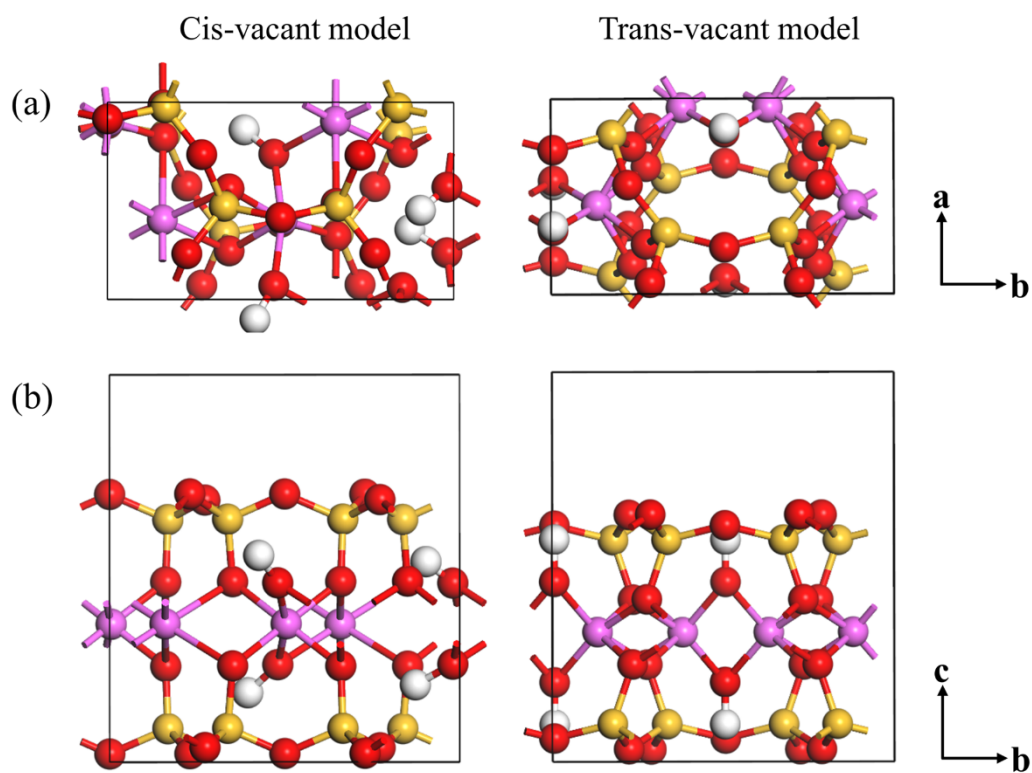


Figure S1. The primitive cell of cv and tv models. Top view (a) and Side view (b). Pink, yellow, red and white spheres represent Al, Si, O and H atoms, respectively.

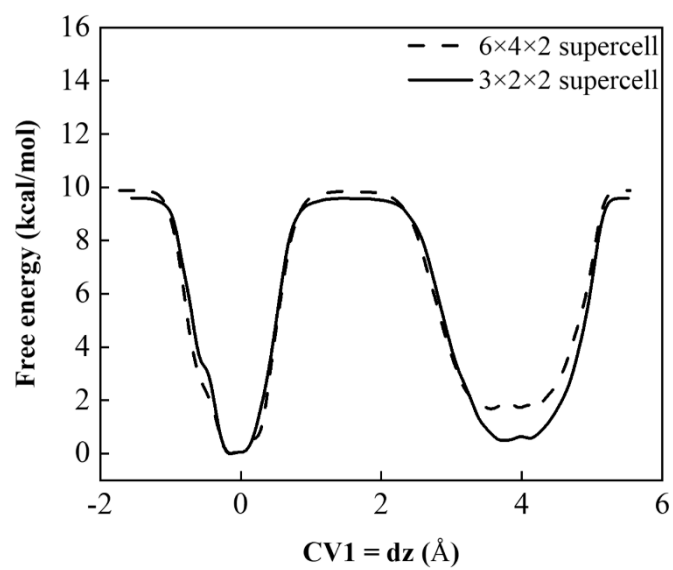
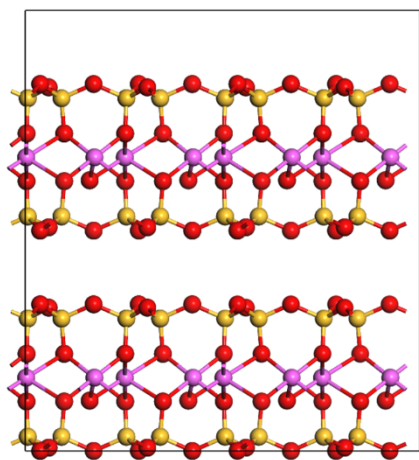
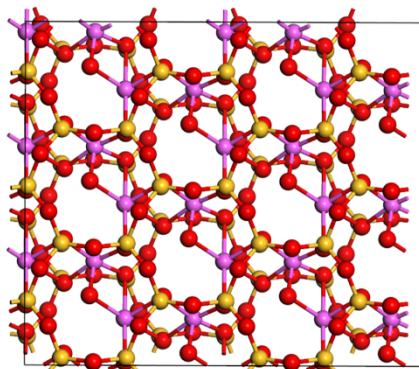


Figure S2. The FES projection on CV1 for the intralayer dehydration reaction simulations of tv smectite with 6×4×2 and 3×2×2 supercells.

Cis-vacant dehydration model



Trans-vacant dehydration model

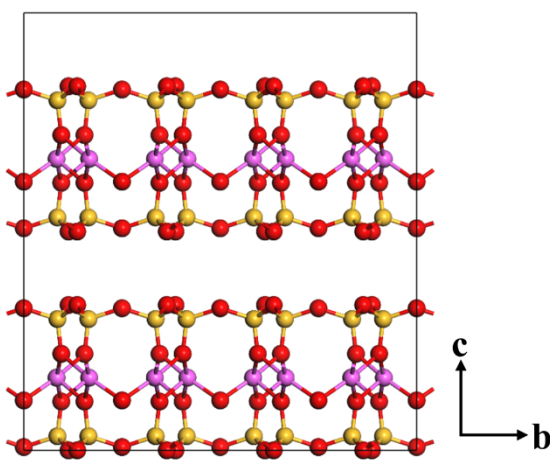
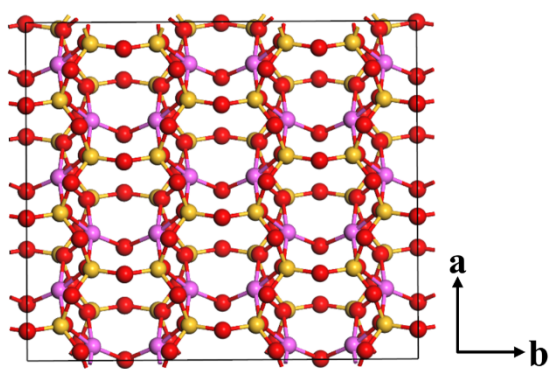


Figure S3. The supercell of *cv* and *tv* dehydration models. Pink, yellow and red spheres represent Al, Si, and O atoms, respectively.

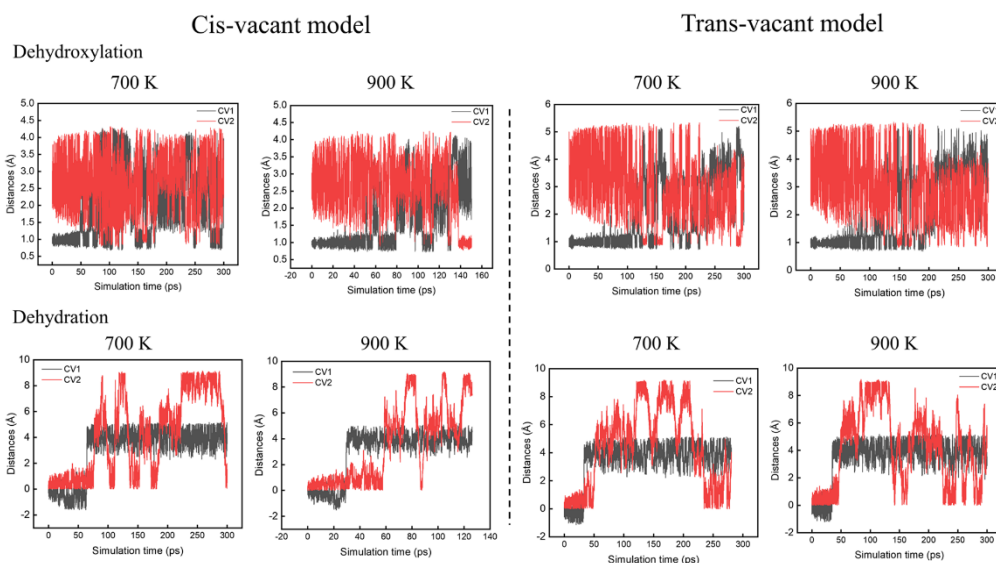


Figure S4. CVs evolutions of the deprotonation reaction simulations of *cv* and *tv* smectites with *cross* mechanism (upper) and the intralayer dehydration reaction (below) at 700 K and 900K.

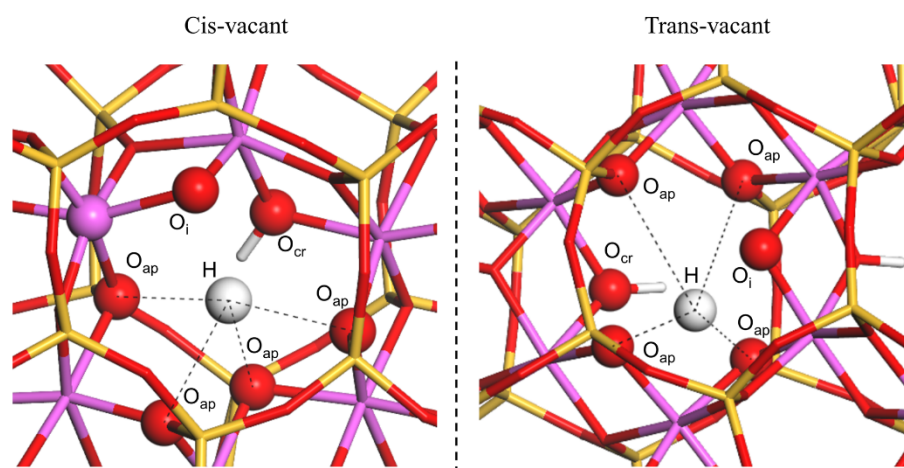


Figure S5. Schematic of protonation/deprotonation occurring at the surrounding apical oxygens of *cv* and *tv* models.

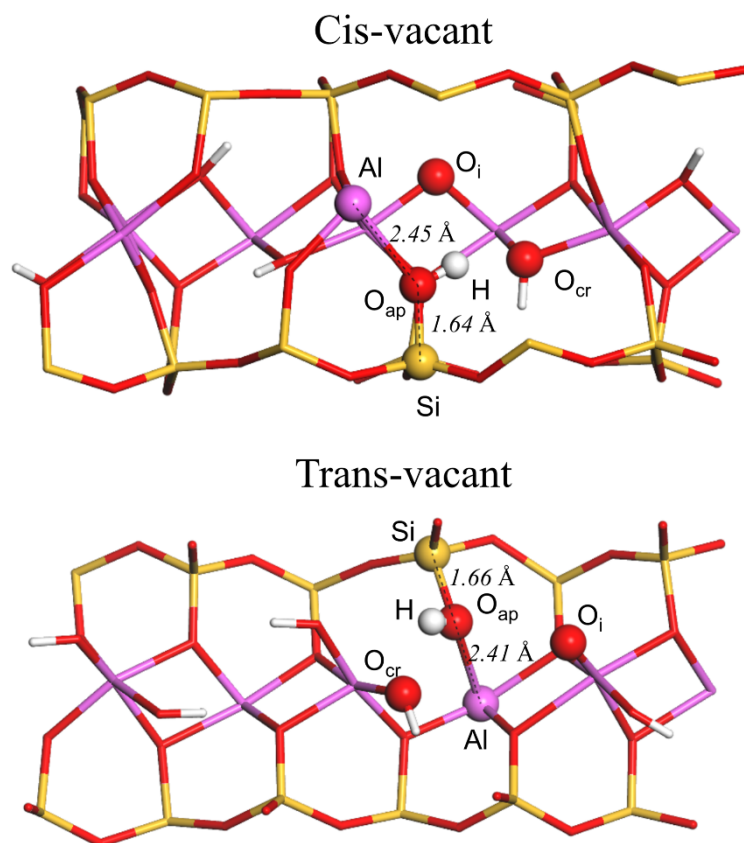
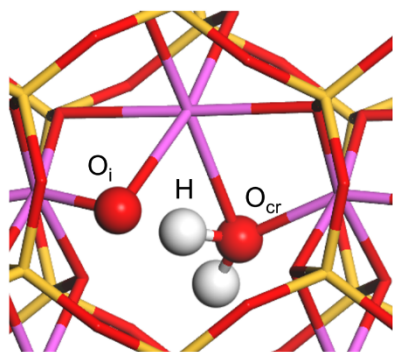


Figure S6. The intermediate structure of the proton coordinated to the O_{ap} of *cv* and *tv* models.

Cis-vacant hydrated intermediate structure



Trans-vacant hydrated intermediate structure

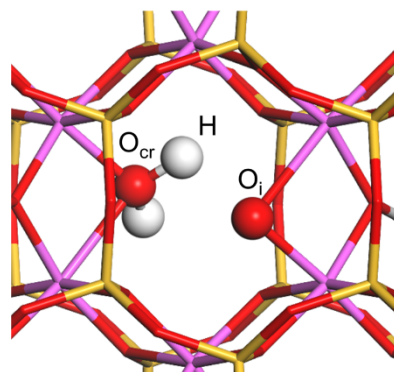


Figure S7. The intralayer hydrated products with water molecules of *cv* and *tv* models.

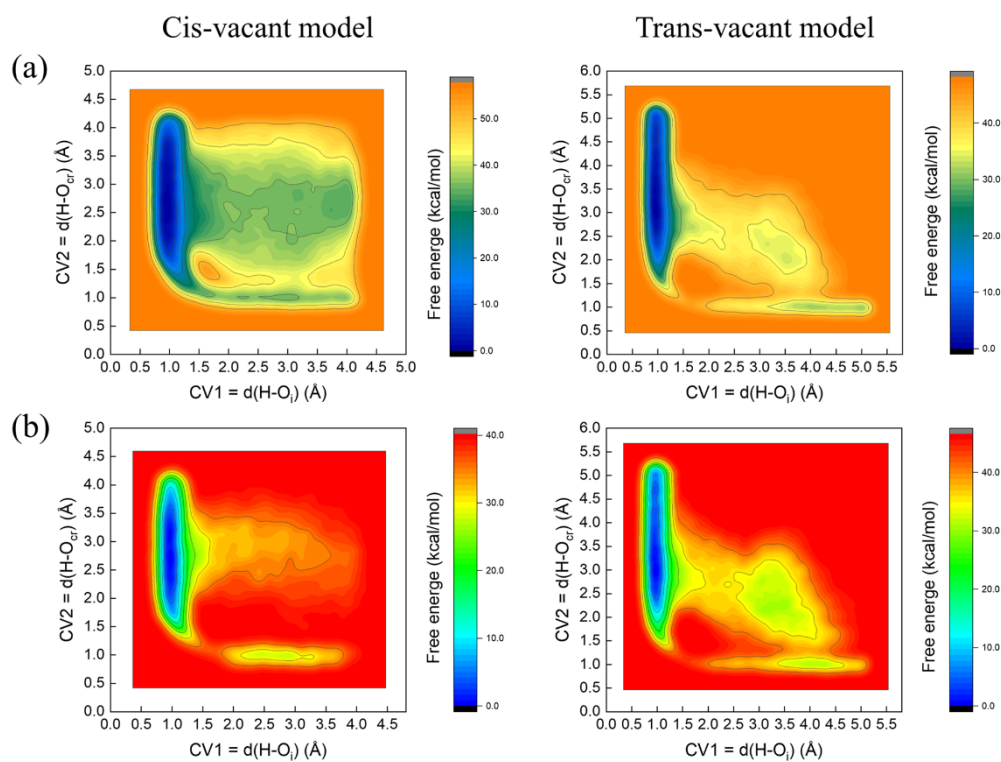


Figure S8. FES for the dehydroxylation reaction simulations of *cv* and *tv* smectites with cross mechanism at 700 K (a) and 900K (b).

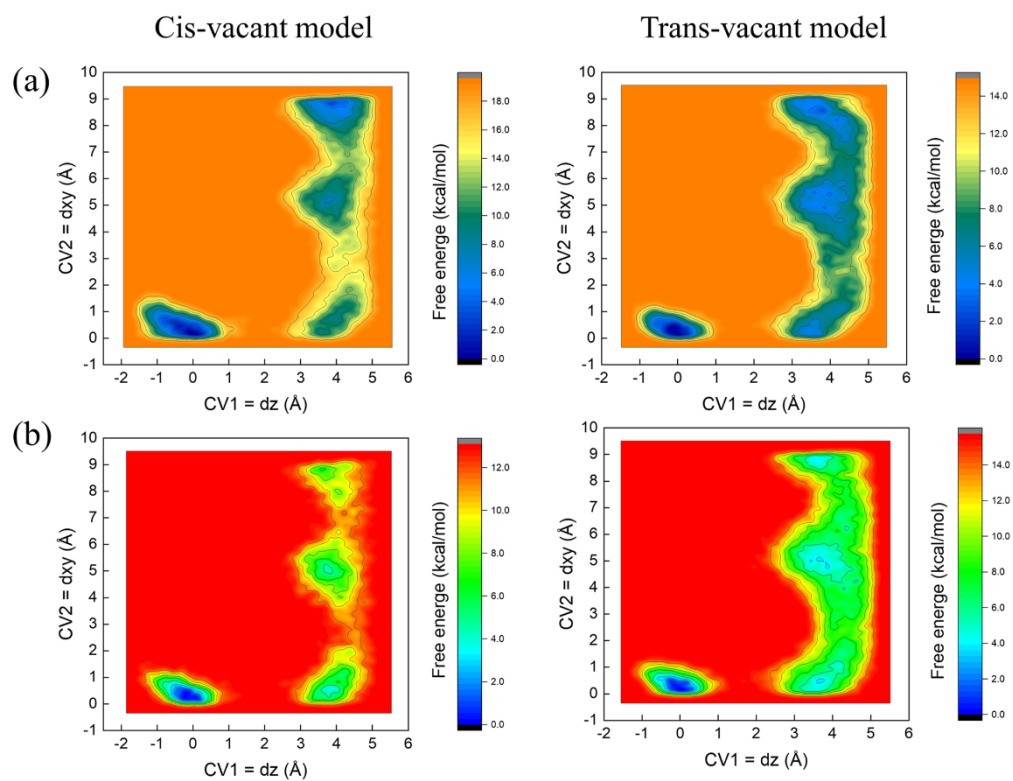


Figure S9. FES for the intralayer dehydration reaction simulations of *cv* and *tv* smectites at 700 K (a) and 900K (b).