

SUPPORTING INFORMATION

Localizing Tetrahedral Aluminum in Nitrate-Bearing Gibbsite to Constrain Defect-Impurity Coupling

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S1. Additional DFT Results

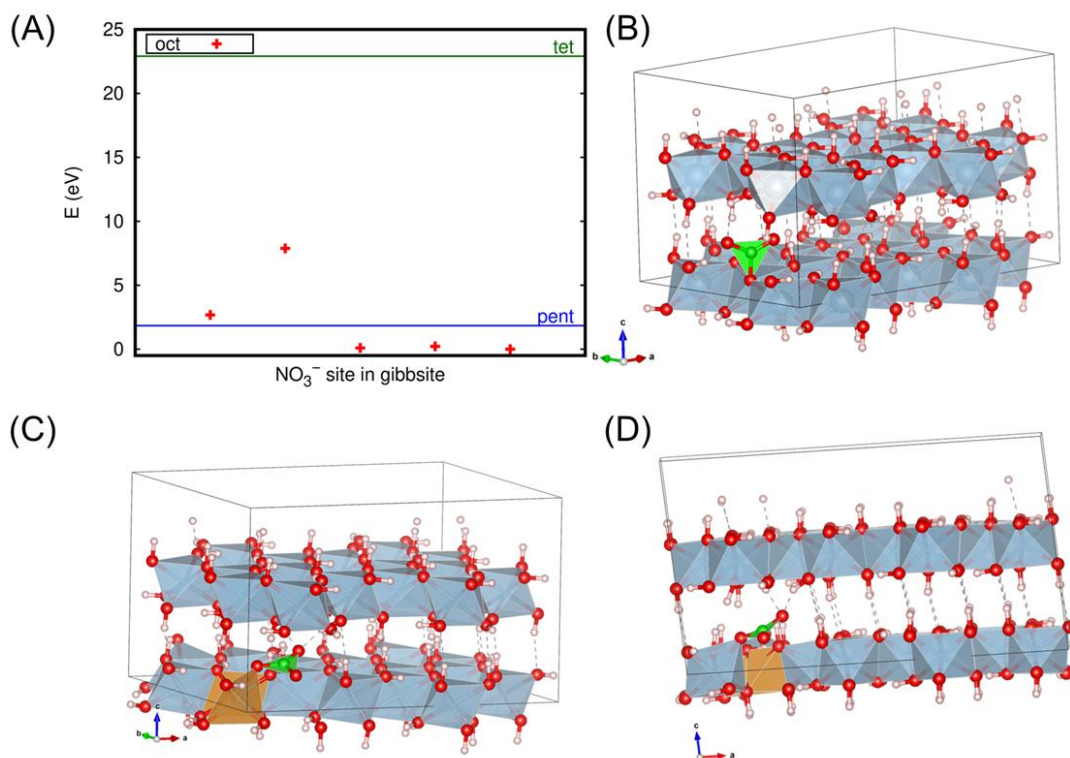


Figure S1. Representative structural models and energetics from DFT simulations.

(A) Relative energies of nitrate substitutions at distinct hydroxyl sites in gibbsite, showing that tetrahedral configurations remain energetically unfavorable compared to higher coordination.

(B) Optimized structure of octahedral gibbsite with a NO₃⁻ group substituting an interlayer OH⁻ from the CONTCAR output. Optimized structure of gibbsite with an NO₃⁻ group substituting an interlayer OH⁻ and (C) breaking the Al-O-Al bridge, leaving two pentahedral coordinated Al sites or (D) one tetrahedral Al site (site 1). In the structural images, hydrogen is shown in pink, oxygen in red, nitrate tetrahedra in green, and undercoordinated Al in orange.

S2. Structural Models (VASP CONTCAR Files)

Al H O

1.0000000000000000

17.3586820016025847 0.0081310804927663 0.2977389459785473
0.0060003771365611 10.2400010316040877 0.0002338828345980
-0.6228128631998289 0.0010573810643942 9.8410980567206252

N O Al H O

1 3 32 95 95

Direct

0.0760099993506270 0.4216592426491557 0.1958937562291966
0.0768513522873108 0.4312399664234249 0.0607102098735574
0.0189841328185510 0.4751368694660675 0.2460977950382484
0.1278772809081218 0.3621577506571281 0.2533698550985254
0.0821112888384154 0.2535506589215979 0.9727693806063129
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0.1712278081109748 0.0191904962093010 0.5089004100799523
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0.3324782555227096 0.4873524851484169 0.9966759830145376
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