Electronic Supplementary Material (ESI) for RSC Advances

Enhanced Li Adsorption and Diffusion in Silicon Nanosheets Based on First Principles Calculations

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Supplementary Information

Computational Details

In our work, silicon nanosheets (SiNSs) are modeled using slab geometry. The initial structure of SiNSs is constructed by cleaving the appropriate surface of the bulk diamond structure of Si. Vacuum spacing of over 12 Å is used in the direction normal to the slab to create isolated slab-boundary conditions. The dangling bonds on the surfaces of SiNSs are passivated with H or F, Cl, I. The typical simulation cell is shown in Figure S1.

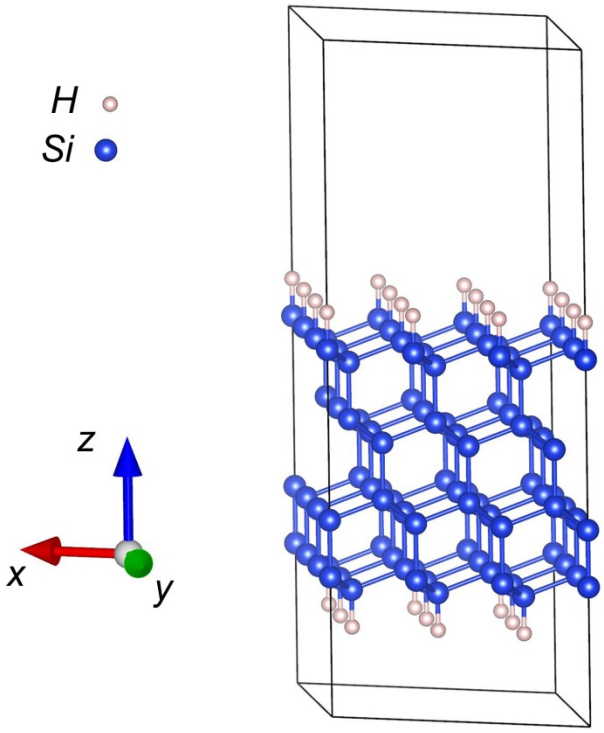

Figure S1. Typical (3×3) simulation cell consisting of 4-layers Si nanosheet slab.

Li surface sites

The most stable positions for Li atom on surface of (111) SiNS are *S1* and *S2* sites (Figure S2). In *S1* site, Li atom sits on the top of a second-layer Si atom. Meanwhile, in *S2* site, Li sits on top of a first-layer Si atom. Both sites have equal Li binding energies with a difference of only 0.002 eV. The minimum Li-Si distances are 2.66 Å.

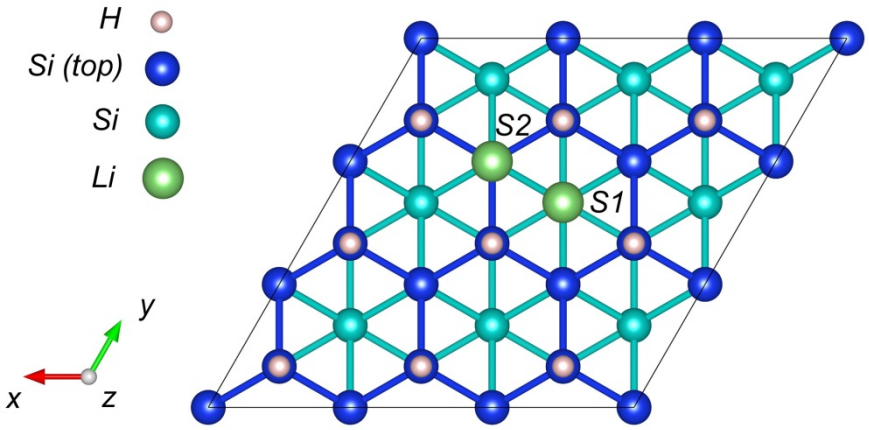

Figure S2. Li surface adsorption sites on (111) SiNS.

Benchmark Calculations

To determine the optimal simulation parameters, we firstly perform a series of benchmark ab initio investigations on Si crystal. To determine the lattice constant in *Quantum Espresso*, the total energy of the given structure is calculated as a factor of the unit cell size. The lattice constant is then defined as a unit cell size, for which the total energy is minimal. We consider the effects of main simulation parameters, such as number of k-points, kinetic energy cutoffs for wavefunctions (ecutwfc) and charge density (ecutrho).

For the diamond crystal structure of Si, the experimentally measured lattice constant is 5.43 Å. Our simulations on Si diamond are performed in cubic simulation cell with 64 atoms. The k-points mesh is varied between 2x2x2, 5x5x5 and 8x8x8; ecutwfc is set as 28 Ry, 32 Ry, 37 Ry and 45 Ry; ecutrho is defined as 320 Ry and 370 Ry.

The obtained results (Figures S3-S4) demonstrate that a change of the simulation parameters (k-point mesh, ecutwfc and ecutrho) affects the numerical values of all total energies, but has no influence on the calculated lattice constants. The results of our calculations are in very good agreement with the experimental values, supporting our choice of pseudopotentials.

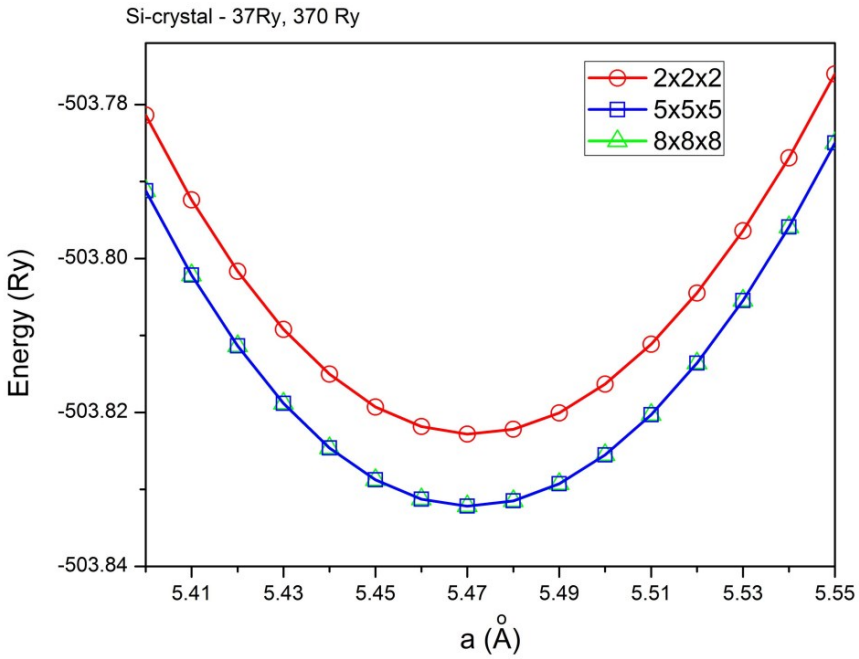

Figure S3. Total energy of Si crystal as a function of k-points mesh. Simulation parameters: ecutwfc=37 Ry, ecutrho=370 Ry.

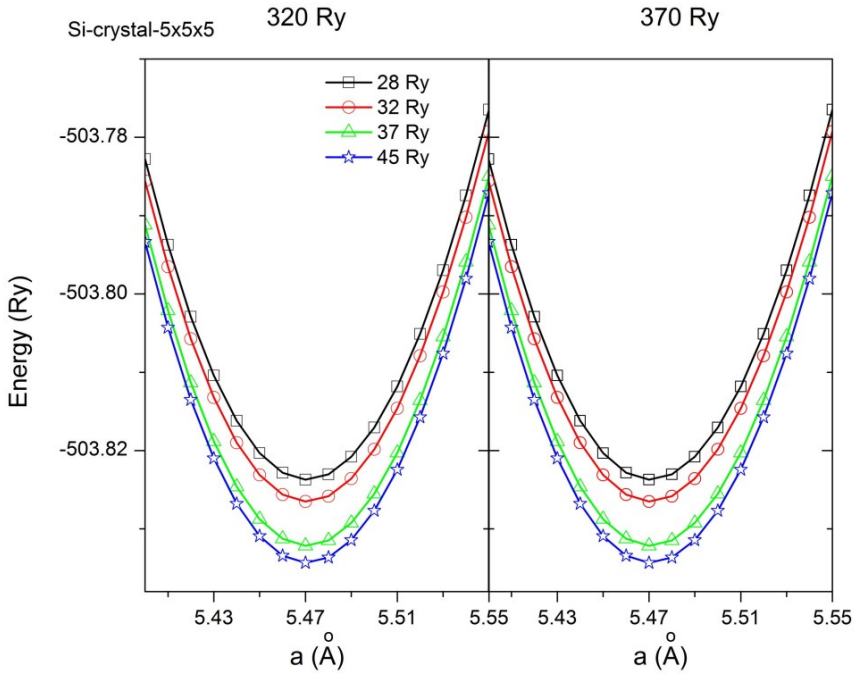

Figure S4. Total energy of Si crystal as a function of kinetic energy cutoffs. Simulation parameters: 5x5x5 k-points mesh.