Supplementary Information (SI) for Physical Chemistry Chemical Physics.
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Supporting Information

---------Calculation details---------

1. Convergence tests

1.1 Cutoff energy convergence tests

Comprehensive cutoff energy convergence tests were performed with the range spanning from 100 Ry to 1000 Ry, covering parameter ranges from extremely low to high precision. The test results are as follows:

Table 1 Cutoff Energy Convergence Tests

CUTOFF (Ry)	Total Energy (Ha)	Energy/Atom (meV)	ΔE (meV/atom)	SCF Steps
100	-617.27400278963296	-131187.5	-	9
200	-617.26790386338689	-131187.5	+1.467	9
300	-617.26834704676696	-131187.5	+0.180	9
400	-617.26702646352032	-131187.6	+0.100	9
500	-617.26722734273699	-131187.5	+0.100	10
600	-617.26706684807493	-131187.5	+0.100	9
700	-617.26713618460712	-131187.5	+0.100	9
800	-617.26706610266262	-131187.5	+0.100	9
900	-617.26710333856295	-131187.5	0.000	9
1000	-617.26705764942744	-131187.5	0.000	9

Scientific Basis for Choosing a Cutoff Energy of 400 Ry

We chose 400 Ry as the working parameter based on the following sufficient scientific evidence:

Extremely early convergence: Starting from 200 Ry, the variation of the total energy is less than 0.2 meV/atom, which is far below the usual convergence criterion in computational materials science (1 meV/atom).

Method robustness: This reflects the excellent numerical stability of the CP2K Gaussian plane - wave hybrid method for the SiC system.

Computational efficiency: On the premise of ensuring accuracy, 400 Ry saves 30-50% of computational resources compared with higher cutoff energies, which is particularly important for our large-scale AIMD simulations.

The values we calculated are far below the widely accepted convergence criterion of 1 meV/atom in the field of computational chemistry, and also meet the more stringent criterion of 0.1 meV/atom. Therefore, the cutoff energy setting of 400 Ry adopted in the calculations is sufficiently converged and reliable, which can ensure the accuracy of all our physical conclusions.

1.2 K-Points convergence tests

Next, fix the cutoff energy at 400 Ry and test the K-point meshes: 1×1×1, 2×2×2, 3×3×3, 4×4×4, 5×5×5. The convergence criterion is set to < 1 eV/atom.

Table 2 K-Points Convergence Tests

K-POINTS	Total Energy (Ha)	Energy/Atom (meV)	ΔE (meV/atom)	SCF Steps
1×1×1/Γ-point	-617.26702646352032	-131220.08	-	9
2×2×2	-617.37290110179606	-131244.47	-24.39	9
3×3×3	-617.37360143980993	-131246.33	-1.86	9
4×4×4	-617.37362835174486	-131246.40	-0.07	9
5×5×5	-617.37362822916884	-131246.40	-0.00	9

1.3 Conclusion

In this CP2K AIMD calculation task, the cutoff energy is 400 Ry and the K-point grid is 3×3×3, which can take into account both high precision and computational efficiency.

2. Spin polarized calculations

Table 3 Final spin polarized

System	MULTIPLICITY
Perfect β-SiC	1
Vacancy-C	3
Vacancy-Si	7
Vacancy-[Si+C]	5

Table 4 Perfect spin polarized

MULTIPLICITY	Total Energy (Ha)	SCF Steps
1	-617.37360143978094	9
2	-617.37360143978094	9
3	-617.37360143978094	9
4	-617.37360143978094	9

Table 5 Vacancy-C spin polarized

MULTIPLICITY	Total Energy (Ha)	SCF Steps
1	-611.50138897572310	10
2	-611.50138897571151	10
3	-611.50138897610896	10
4	-611.50138897558531	10
5	-611.50138897561919	10

6	-611.50138897581280	10
7	-611.50138897562158	10
8	-611.50138897541717	10
9	-611.50138897575482	10
10	-611.50138897607894	10

Table 6 Vacancy-Si spin polarized

MULTIPLICITY	Total Energy (Ha)	SCF Steps
1	-613.11911497721746	15
2	-613.11911497742756	15
3	-613.11911497723099	15
4	-613.11911497743279	15
5	-613.11911497740664	15
6	-613.11911497709957	15
7	-613.11911497779568	15
8	-613.11911497701885	15
9	-613.11911497755398	15
10	-613.11911497752760	15

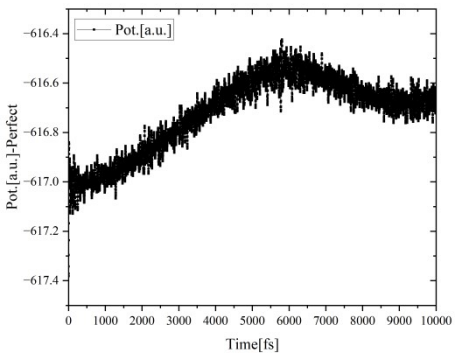
Table 7 Vacancy-[Si+C] spin polarized

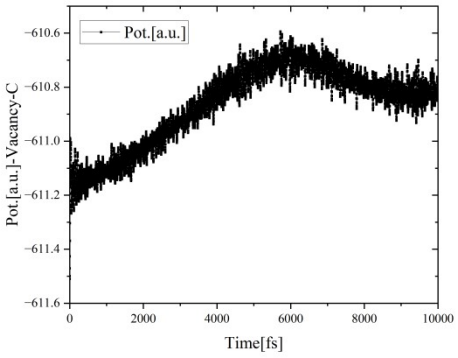
MULTIPLICITY	Total Energy (Ha)	SCF Steps
1	-607.27707249745708	20

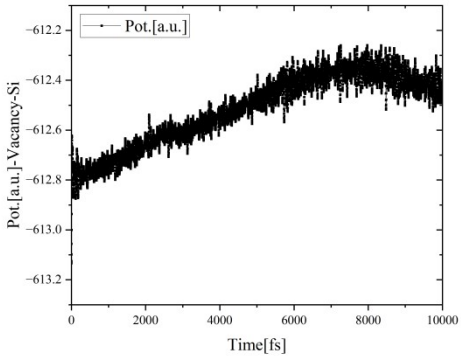
2	-607.27707249760351	20
3	-607.27707249725802	20
4	-607.27707249759499	20
5	-607.27707249789057	20
6	-607.27707249767832	20
7	-607.27707249766092	20
8	-607.27707249731554	20
9	-607.27707249768127	20
10	-607.27707249765126	20

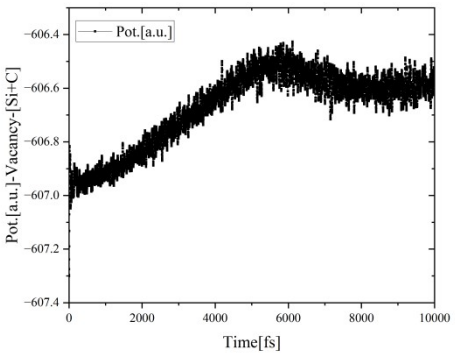
3. AIMD Calculation

Pot









Temperature