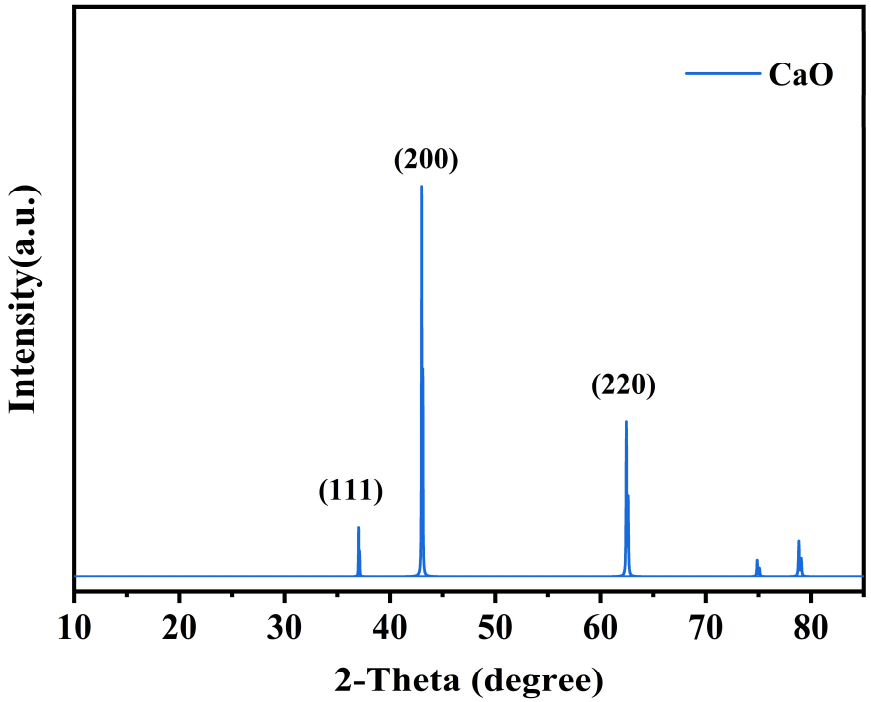


Figure S1. XRD calculated patterns of CaO crystals.

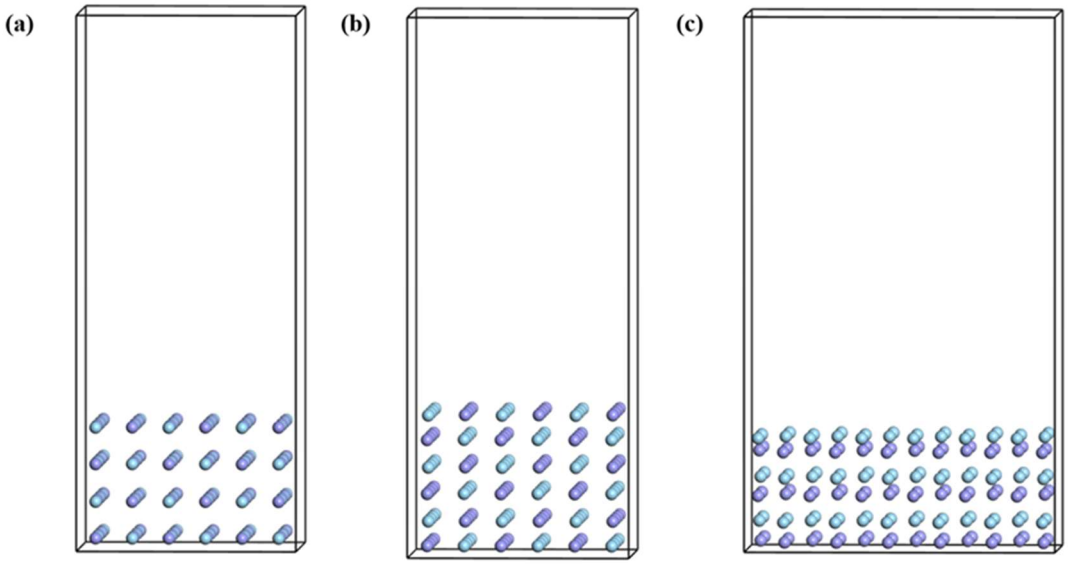


Figure S2. Different crystal face models of CaO: (**a**) CaO(100), (**b**) CaO(110) and (**c**) CaO(111) (Ca and O atoms are represented by purple and blue spheres, respectively).

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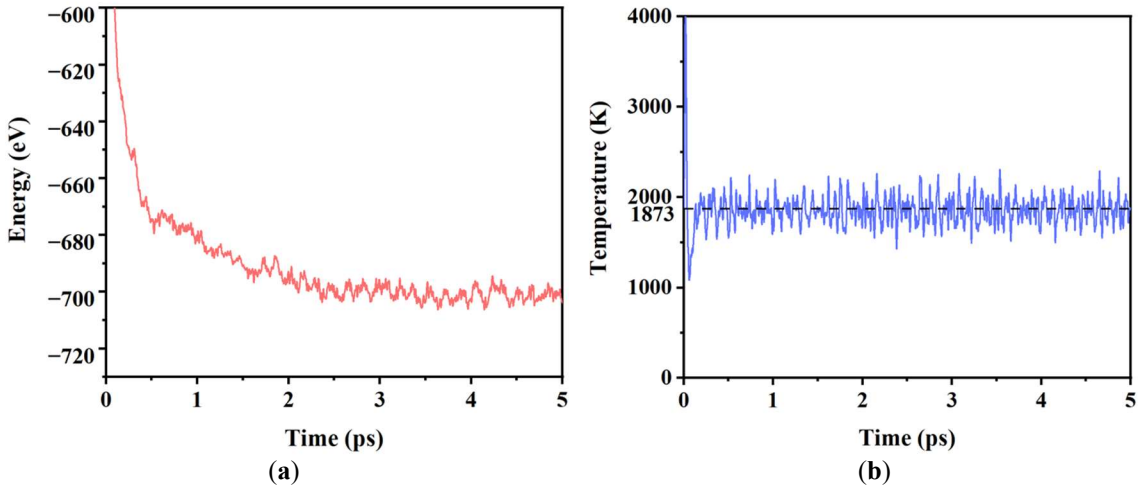


Figure S3. Variations of system temperature (**a**) total energy of C_3S and C_2S melt at 1873 K; (**b**) during AIMD simulations of C_3S and C_2S melt at 1873 K.

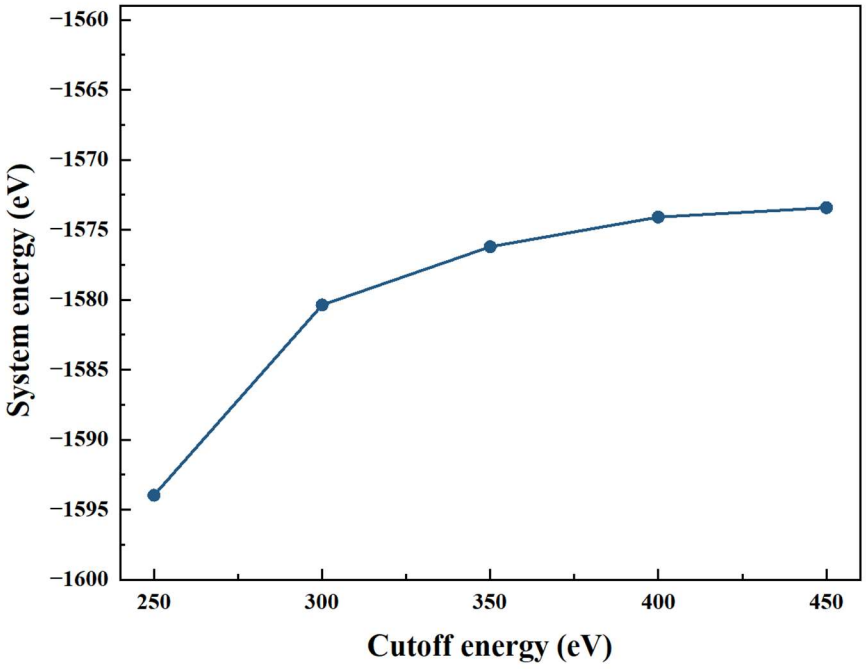


Figure S4. Cutoff-energy convergence test for the C_3S/CaO(100) system.

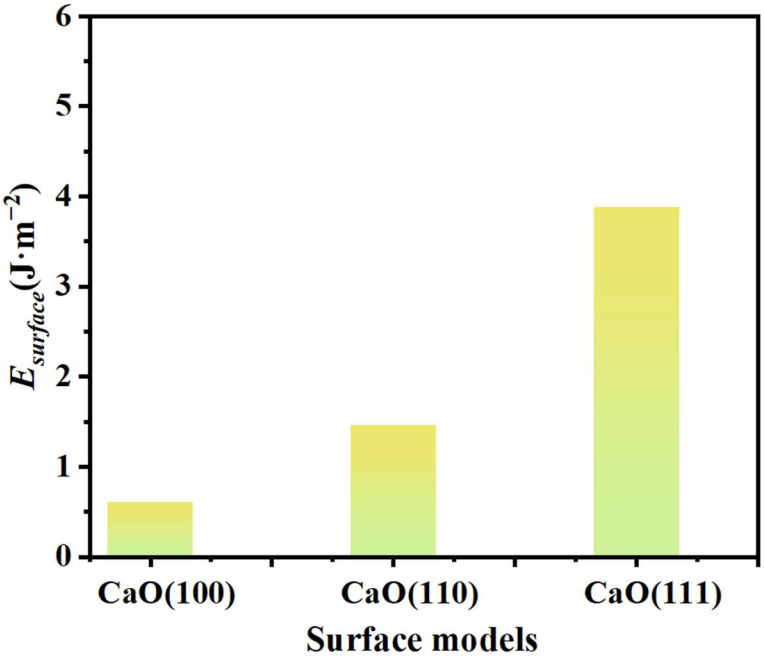


Figure S5. Surface energies of different crystal faces of CaO.

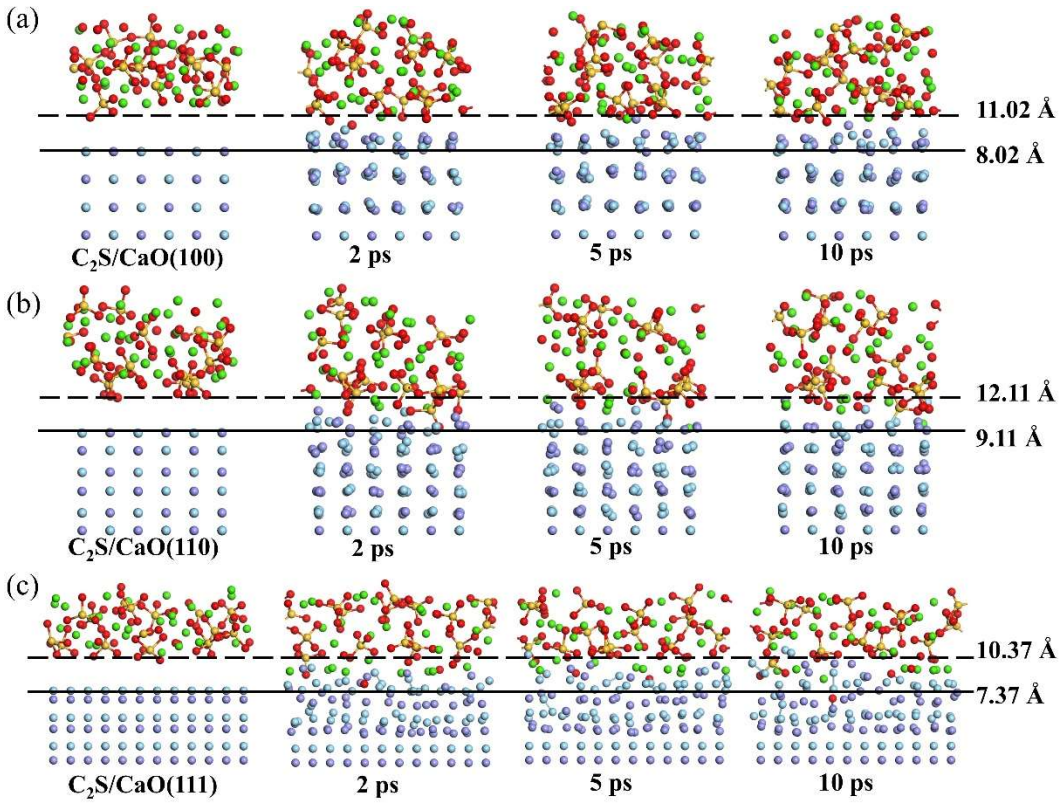


Figure S6. Atomic structural evolution of the C_2S/CaO interface: (**a**) C_2S/CaO(100), (**b**) C_2S/CaO(110) and (**c**) C_2S/CaO(111)(The solid and dotted lines represent the upper surface of the CaO and the lower surface of C_2S in the initial interface model, respectively; Ca, Si and O atoms in the C_2S are represented by green, yellow and red spheres, respectively; Ca and O atoms in CaO are represented by purple and blue spheres, respectively).

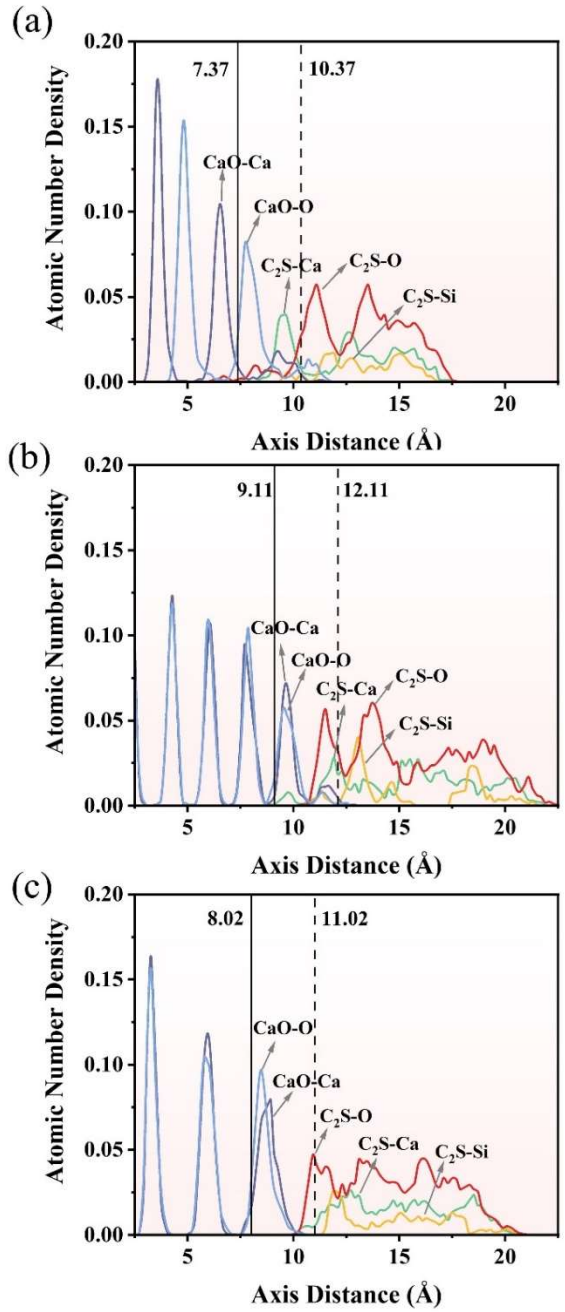


Figure S7. Atomic number density distribution of the C_2S/CaO interface: (**a**) C_2S/CaO(100), (**b**) C_2S/CaO(110) and (**c**) C_2S/CaO(111) (The solid and dotted lines are the upper surface of the CaO and the lower surface of C_2S in the initial interface model, respectively).

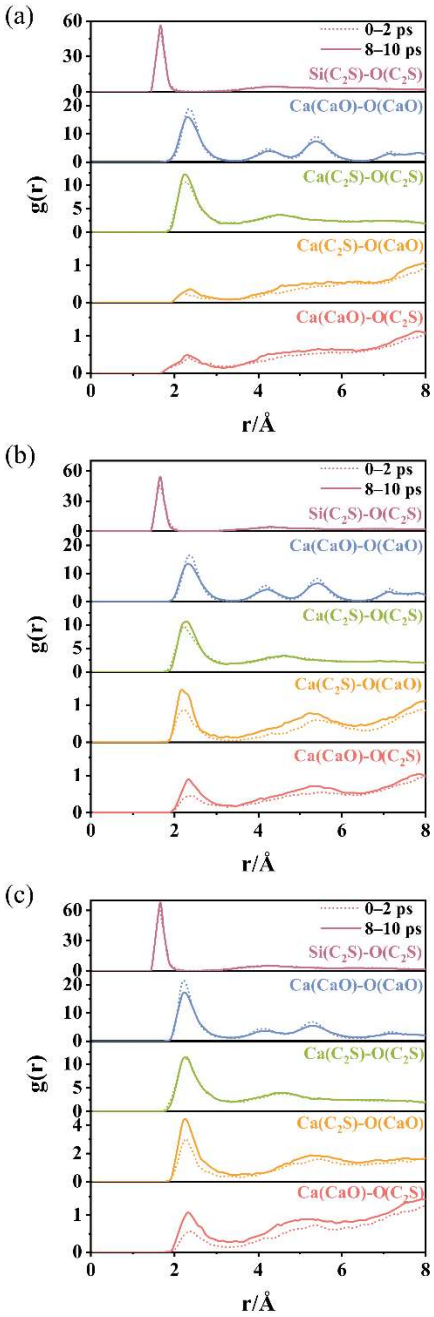


Figure S8. Radial distribution functions of the C_2S/CaO interface at the initial (0-2 ps, dotted lines) and final (8-10 ps, solid lines) stages of the simulated reaction: (**a**) C_2S/CaO(100), (**b**) C_2S/CaO(110) and (**c**) C_2S/CaO(111).

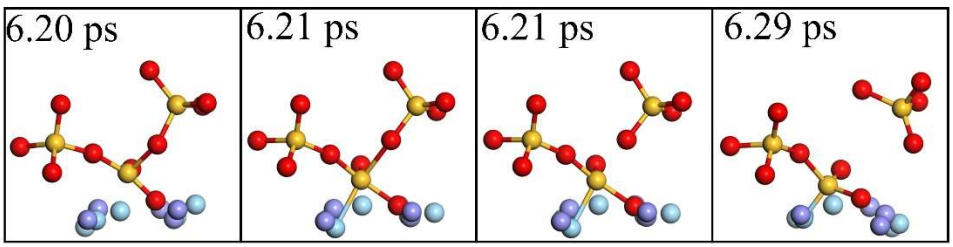


Figure S9. Interaction mechanism between Si atoms in C_2S and CaO(110) (Ca and O atoms in CaO are represented by purple and blue spheres, respectively; Si and O atoms in C_2S are represented by yellow and red spheres, respectively).

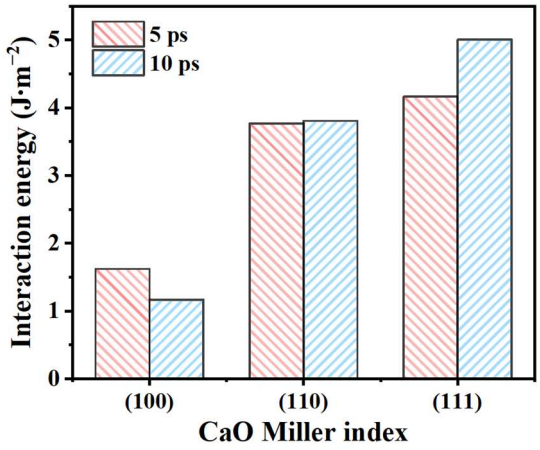


Figure S10. Interaction energies of C_2S and CaO surface with different Miller indices.

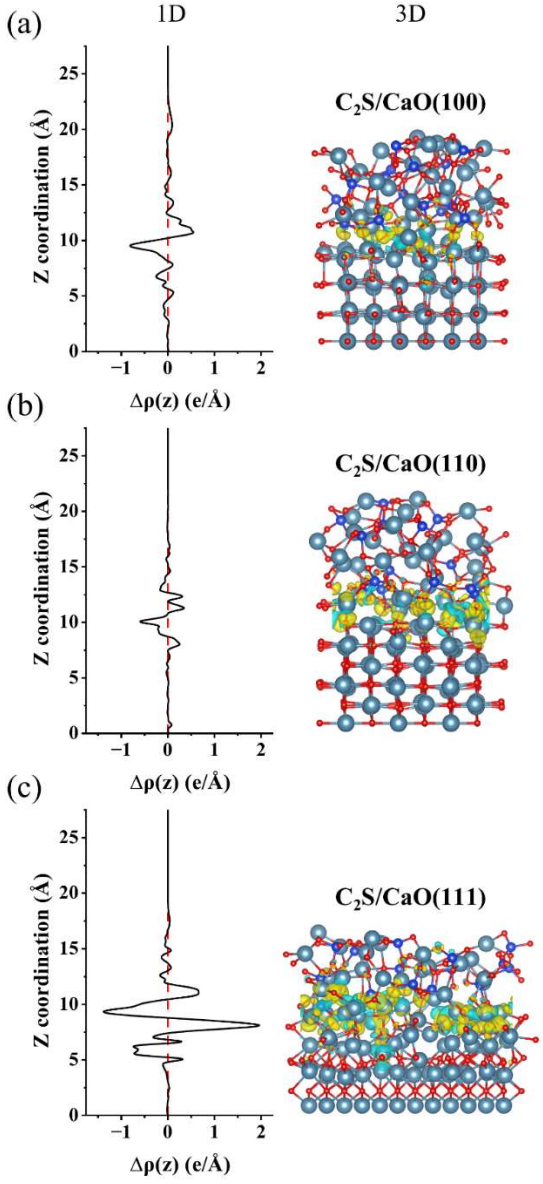


Figure S11. One-dimensional and three-dimensional differential charge density of the C_2S/CaO interface: (**a**) C_2S/CaO(100), (**b**) C_2S/CaO(110) and (**c**) C_2S/CaO(111) (Si, Ca and O atoms are represented by blue, gray and red balls respectively; yellow and cyan represent the regions of electron accumulation and loss respectively; the isosurface value is 0.005 e/bohr^3).

Table S1. The Optimized and experimental lattice parameters.

Crystal Type	Unit Cell Parameters	Data in This Paper	Comparative Data [40]
CaO	a/Å	4.79	4.81
	b/Å	4.79	4.81
	c/Å	4.79	4.81

Table S2. Atomic counts in the simulated systems of C_3S and C_2S.

	Ca	Si	O	SUM	g/cm^3
C_3S	36	12	60	108	3.00
C_2S	30	15	60	105	3.07

Table S3. Average Bader charge values of atoms at the C_3S/CaO interface (Positive values denote electron loss, negative values denote electron gain).

Model	C_3S (e)			CaO (e)	
	Ca	Si	O	Ca	O
C_3S/CaO(100)	1.36	2.12	−1.25	1.32	−1.31
C_3S/CaO(110)	1.37	2.10	−1.27	1.29	−1.27
C_3S/CaO(111)	1.38	2.41	−1.22	1.16	−1.23

Table S4. Bader charges of C_2S/CaO interface (Positive values denote electron loss, negative values denote electron gain).

Model	C_2S (e)	CaO (e)
C_2S/CaO(100)	−1.91	1.91
C_2S/CaO(110)	−0.70	0.70
C_2S/CaO(111)	3.07	−3.07

Table S5. Average Bader charge values of atoms at the C_2S/CaO interface (Positive values denote electron loss, negative values denote electron gain).

Model	C_2S (e)			CaO (e)	
	Ca	Si	O	Ca	O
C_2S/CaO(100)	1.40	2.24	−1.29	1.32	−1.30
C_2S/CaO(110)	1.39	2.27	−1.28	1.30	−1.29
C_2S/CaO(111)	1.43	2.17	−1.20	1.16	−1.21