Table S1. Calculation details and the surface energy of pristine low-index surfaces of calcium silicates.

Calcium Silicate Phase	Low-Index Surfaces	Slab Model Size			*n*	Brillouin Zone Mesh	Surface Energy (J/m²)
		a (Å)	b (Å)	c (Å)			
β-C_2S	(100)	6.76	9.30	42.39	5	3 × 2 × 1	0.84
	(010)	9.30	5.53	42.23	4	2 × 4 × 1	0.96
	(001)	5.53	6.76	41.90	3	4 × 3 × 1	0.96
	(110)	9.30	8.73	42.54	6	2 × 2 × 1	1.14
	(101)	11.23	6.76	41.67	6	2 × 3 × 1	0.98
	(011)	5.53	11.50	41.98	5	4 × 2 × 1	1.00
	(111)	8.73	11.23	41.30	7	3 × 2 × 1	1.38
M3-C_3S	(100)	7.11	12.16	39.52	3	4 × 2 × 1	1.32
	(010)	11.53	9.28	43.28	4	2 × 3 × 1	1.00
	(001)	9.28	7.11	47.33	3	3 × 4 × 1	0.95
	(110)	12.16	11.69	43.81	5	2 × 2 × 1	1.02
	(101)	18.30	7.11	44.32	5	2 × 4 × 1	1.35
	(011)	13.55	9.28	44.59	5	2 × 3 × 1	1.30
	(111)	11.69	14.09	40.98	6	2 × 2 × 1	1.08

Note: *n* is the number of formula units contained in the slab model, surface energy is calculated using $\text{surface energy} = (E_S - n \times E_B) / (2 \times A)$, where E_S is the total energy of the slab model, E_B is the total energy per formula unit of the bulk structure, and A is the surface area of the slab.

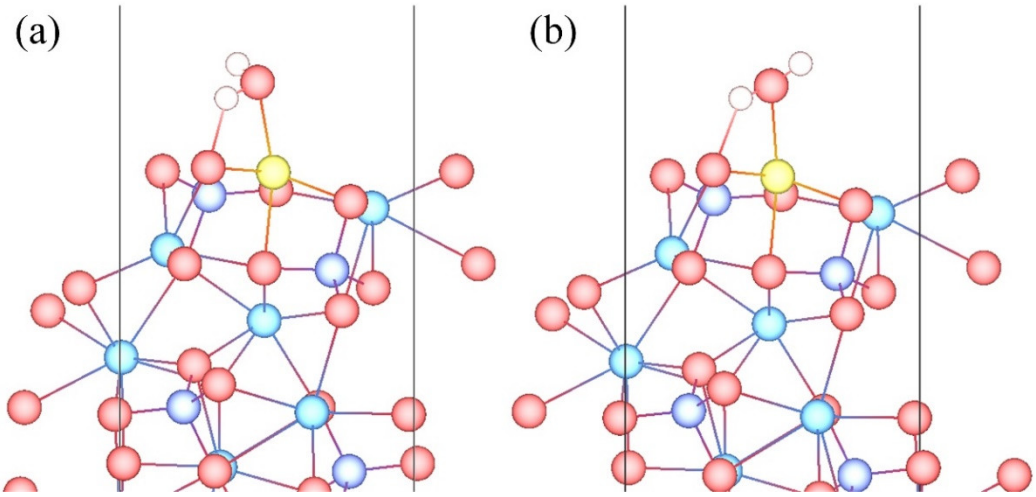

Figure S1. Water adsorption configuration on β-C_2S (001) surface: (**a**) −0.994 eV, and (**b**) −0.915 eV. Please refer to Figure 1 for the atom type.