Supplementary Information

Unveiling the heavy-metal ion critical role in γ-dicalcium silicate: From solidification to early hydration

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Table S1. Crystal structure parameters for each configuration following geometry optimization

		Volume/Å^3	a /Å	b /Å	c/Å	Alpha/°	beta/°	gamma/°
Prue		1582.41	10.25	11.34	13.62	90.00	90.00	90.00
Cd	Ca1	1581.53	10.25	11.35	13.62	90.05	89.96	89.96
	Ca2	1580.17	10.25	11.32	13.62	90.00	90.00	90.00
	Si	1598.19	10.33	11.36	13.63	90.00	90.00	90.09
	Si+O	1601.20	10.32	11.37	13.65	89.64	89.71	90.15
	V	1606.79	10.29	11.45	13.64	89.98	90.09	90.15
Ba	Ca1	1598.53	10.29	11.35	13.69	89.64	90.22	90.24
	Ca2	1602.66	10.25	11.40	13.71	90.00	90.00	90.08
	Si	1607.10	10.38	11.38	13.60	90.00	90.00	89.89
	Si+O	1603.95	10.33	11.36	13.67	89.54	89.98	90.16
	V	1617.83	10.28	11.51	13.69	89.65	90.35	90.55
Cr	Ca1	1574.30	10.22	11.36	13.56	90.22	90.13	90.16
	Ca2	1570.95	10.25	11.27	13.61	90.00	90.00	90.29
	Si	1587.51	10.27	11.35	13.62	90.00	90.00	89.99
	Si+O	1589.55	10.27	11.35	13.64	89.94	89.87	90.09
	V	1600.35	10.27	11.42	13.66	90.04	89.99	90.02

Table S2. Bond lengths and angles of adsorbed water molecules on surfaces

Site	H_W−O_S/Å	Ca−O_W/Å	H_W−O_W/Å	∠HOH/deg
Pure-I	1.47	2.56	1.07	103.50
Pure-II	1.47	2.42	1.07	108.38
Pure-III	1.69	2.46	1.01	96.15
Pure-IV	0.97	2.35	2.55	/
Cr-I	1.49	2.40	1.06	110.25
Cr-II	1.52	2.40	1.06	109.96
Cr-III	1.68	2.46	1.01	96.10
Cr-IV	1.81	2.49	1.00	95.17

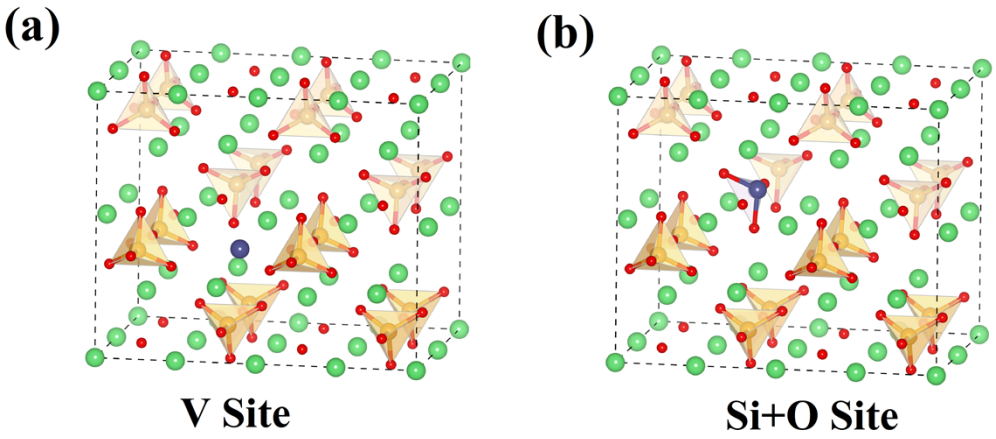

Figure S1. Schematic structures: **(a)** Ca substitution site and, **(b)** Si+O substitution site

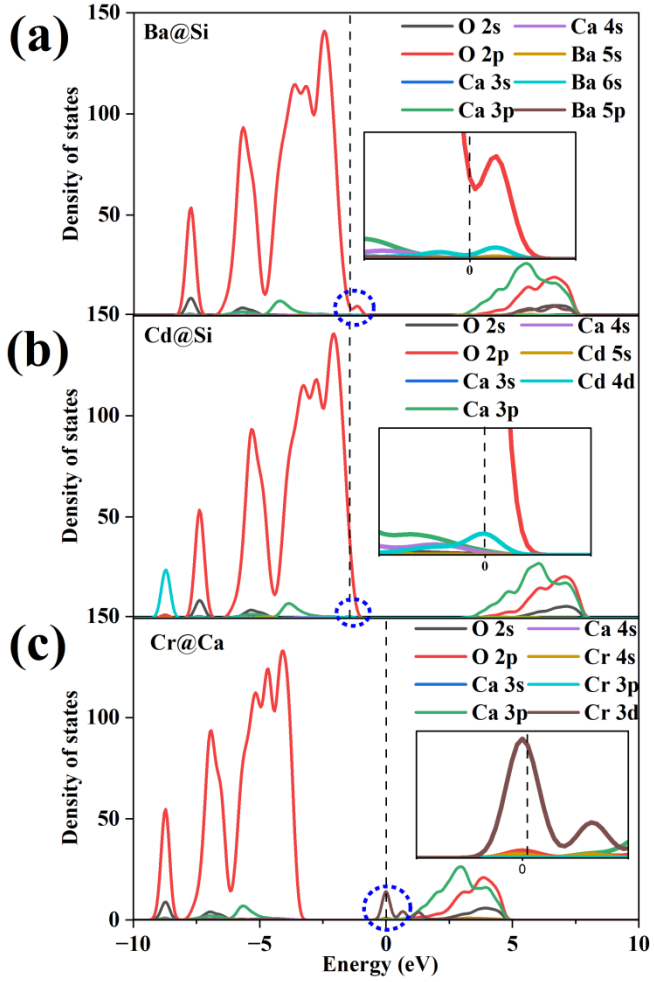

Figure S2. **Density of states (DOS)**: **(a)** Ba-doped γ-C_2S, **(b)** Cd-doped γ-C_2S in Si site, **(c)** Cr-doped γ-C_2S in Ca site.

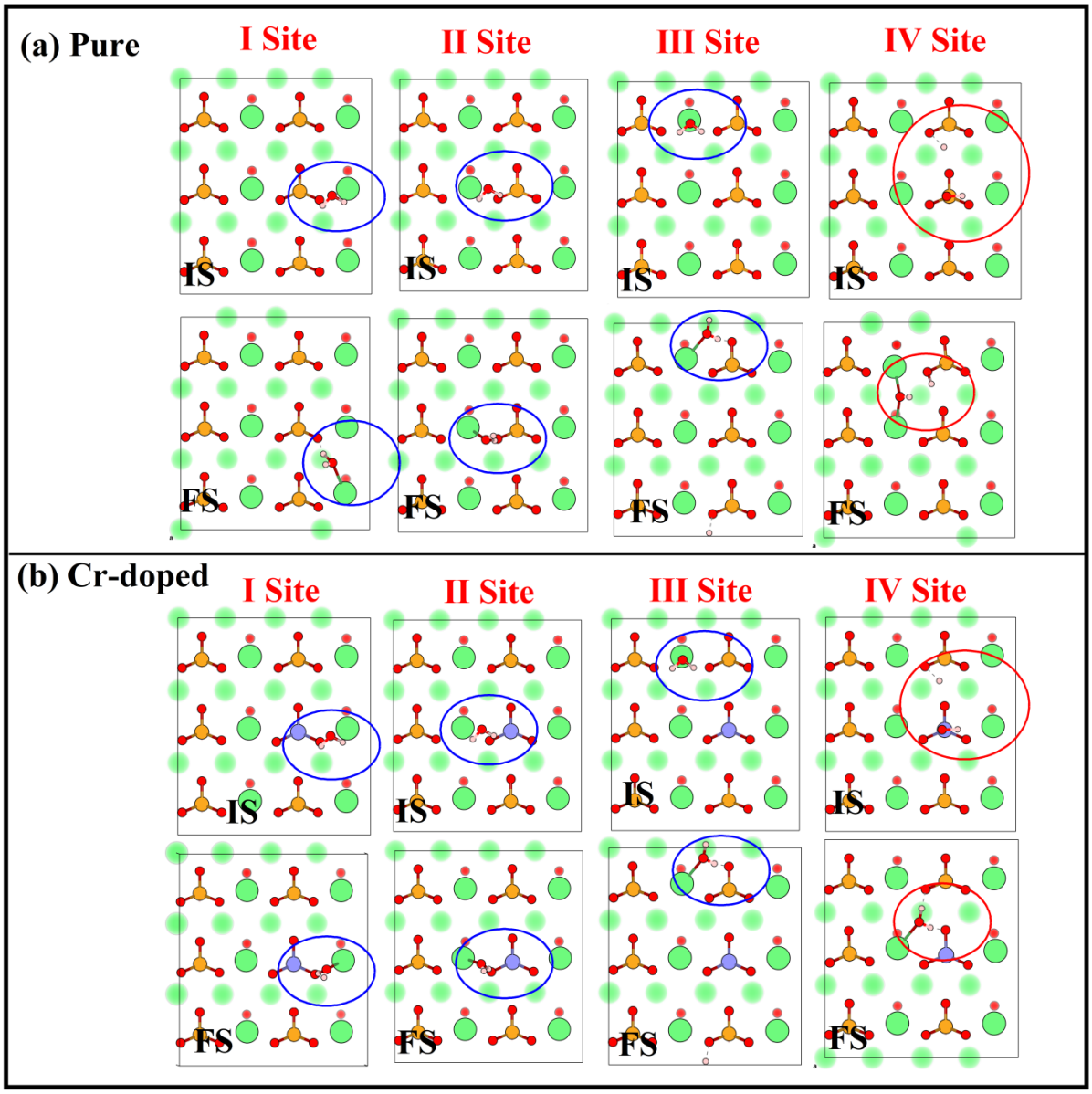

Figure S3. Water adsorption initial (IS) and final state (FS) configurations: (a) pure γ-C_2S, (b) Cr-doped γ-C_2S

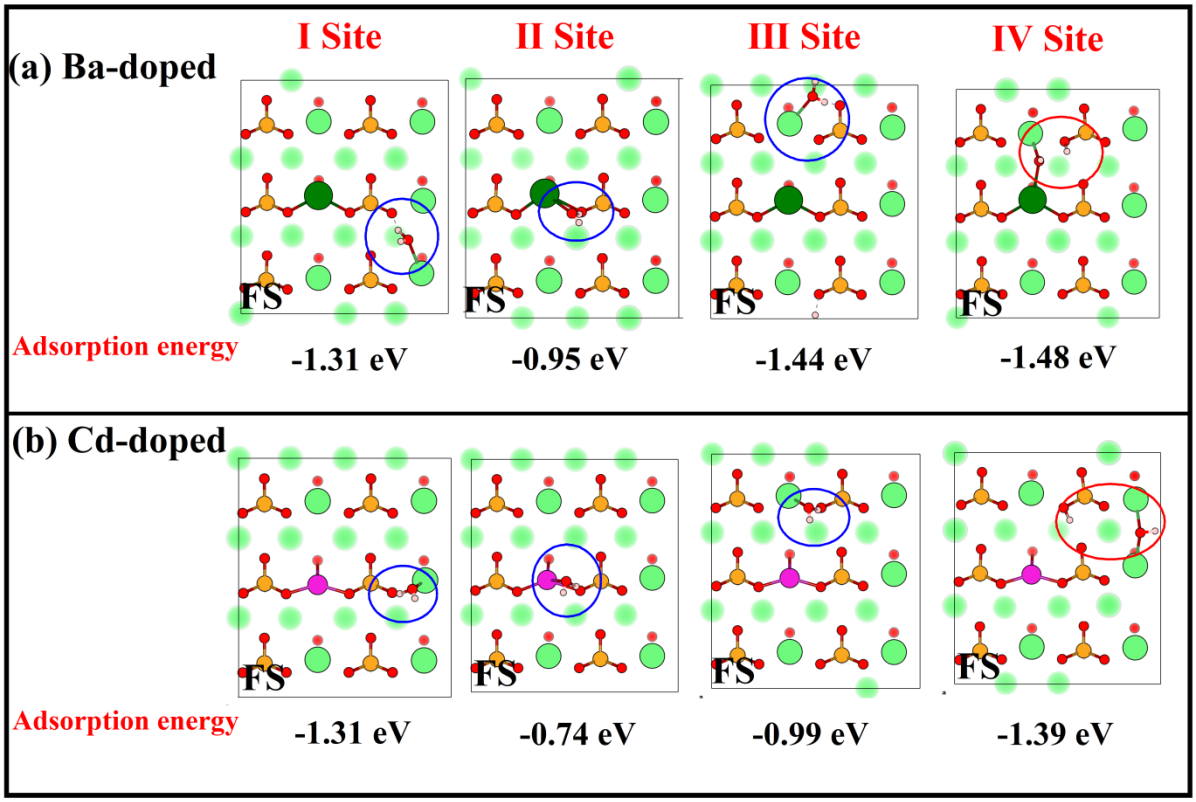

Figure S4. Water adsorption final state (FS) configurations: (a) Ba-doped, (b) Cd-doped γ-C_2S