

Synergistic adsorptions of Na_2CO_3 and Na_2SiO_3 on calcium minerals revealed by spectroscopic and *ab initio* molecular dynamics studies

Supplementary Information

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Table S1. Atomic concentrations (at.%) and binding energies (BE) of the species identified by XPS onto CaF₂ with different treatments. L=Low; H=High.

Elements	CaF ₂		CaF ₂ + Na ₂ CO ₃		CaF ₂ + Na ₂ CO ₃		CaF ₂ + Na ₂ SiO ₃		CaF ₂ + Na ₂ SiO ₃		CaF ₂ + Na ₂ CO ₃ + Na ₂ SiO ₃		CaF ₂ + Na ₂ CO ₃ + Na ₂ SiO ₃		
	-		5.10 ⁻³ mol.L ⁻¹		5.10 ⁻² mol.L ⁻¹		5.10 ⁻³ mol.L ⁻¹		5.10 ⁻² mol.L ⁻¹		5.10 ⁻³ mol.L ⁻¹		5.10 ⁻² mol.L ⁻¹		
	BE	at.%	BE	at.%	BE	at.%	BE	at.%	BE	at.%	BE	at.%	BE	at.%	
Na(1s)	-	-	1071.6	3.5	1071.4	4.5	1071.9	0.8	1071.8	4.2	1071.3	5.0	1071.2	11.9	
F(1s)	685.2	34.7	685.1	35.9	684.8	28.7	685.3	33.6	685.0	9.9	685.0	16.7	684.8	4.0	
O(1s)		14.5		18.3		16.8		22.9		43.8		34.0		42.6	
	<i>H₂O</i>	533.2	3.4	533.0	3.8	-	-	-	-	-	-	-	-	-	
	<i>O-C (CO₃)</i>	532.1	11.1	531.9	14.5	531.7	15.9	531.2	2.5	531.0	4.3	531.6	8.7	530.9	14.3
	<i>O-C (HCO₃)</i>	-	-	-	-	533.6	0.8	-	-	-	-	533.6	4.1	533.5	1.0
	<i>O-Si</i>	-	-	-	-	-	-	532.5	20.5	532.6	39.4	532.5	21.2	532.2	27.3
Ca(2p)		20.1		20.2		17.0		19.7		6.8		12.7		3.6	
	<i>2p_{1/2}</i>	351.8	-	351.7	-	351.3	-	351.8	-	351.5	-	351.7	-	351.3	-
	<i>2p_{3/2}</i>	348.2	-	348.1	-	347.8	-	348.2	-	348.0	-	348.2	-	347.8	-
C(1s)		26.2		18.7		29.5		16.4		19.2		24.0		27.7	
	<i>C-C/C-H</i>	284.6	21.7	284.6	13.7	284.6	24.2	284.6	13.0	284.6	11.7	284.6	12.0	284.6	17.8
	<i>C-O</i>	285.8	2.1	286.1	2.5	286.0	2.0	285.9	2.0	286.3	4.5	285.6	6.0	286.2	2.5
	<i>C=O</i>	286.7	1.3	288.2	1.1	288.3	1.3	287.1	0.8	288.0	1.3	287.1	2.6	288.2	1.3
	<i>CO₃</i>	288.8	1.1	289.4	1.5	289.3	2.0	289.1	0.6	289.3	1.7	289.3	3.4	289.3	6.2
Si(2p)		3.0		1.7		2.7		5.7		16.1		5.3		10.2	
	<i>HBE</i>	103.1	0.4	102.9	0.5	102.8	0.6	103.2	4.5	103.2	16.1	103.3	3.9	103.1	5.6
	<i>LBE</i>	102.0	2.6	101.8	1.3	101.8	2.1	102.1	1.2	-	-	102.3	1.4	102.2	4.6

Table S2. Ratio of atomic concentration of the elements measured by XPS on CaF₂ with different treatments.

	CaF ₂	CaF ₂ + Na ₂ CO ₃	CaF ₂ + Na ₂ CO ₃	CaF ₂ + Na ₂ SiO ₃	CaF ₂ + Na ₂ SiO ₃	CaF ₂ + Na ₂ CO ₃ + Na ₂ SiO ₃	CaF ₂ + Na ₂ CO ₃ + Na ₂ SiO ₃
	-	5.10 ⁻³ mol.L ⁻¹	5.10 ⁻² mol.L ⁻¹	5.10 ⁻³ mol.L ⁻¹	5.10 ⁻² mol.L ⁻¹	5.10 ⁻³ mol.L ⁻¹	5.10 ⁻² mol.L ⁻¹
F/Ca	1.73	1.78	1.68	1.70	1.47	1.32	1.12
Na/Carb	N/A	2.40	2.25	N/A	N/A	N/A	N/A
O/Si	N/A	N/A	N/A	3.61	2.45	4.03	2.67
O/Na	N/A	4.17	3.55	24.35	9.30	N/A	N/A

Figure S1. XPS O(1s) spectra of CaF₂ conditioned in deionized water (a), in a 5.10⁻³ mol.L⁻¹ Na₂CO₃ solution (orange), and in a 5.10⁻² mol.L⁻¹ Na₂CO₃ solution (red) (b), and in a 5.10⁻³ mol.L⁻¹ (light blue), and a 5.10⁻² mol.L⁻¹ (dark blue) Na₂SiO₃ solutions (c). For each XPS spectra, the different components and their attributions are exhibited.

