Supporting information

A facile recrystallization strategy for fabrication of nanocrystalline microspheres of sulfatic sodalite with high thermal stability

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Table S1 Sample codes, synthesis conditions, Si/Al ratios and phases

Sample Codes	pH of gel	Temp. (℃)	Time (h)	Si/Al ratio	Phases
SOD-1	/	200	4	1.06	B-sodalite
gel-4	4	/	/	0.96	thenardite (Na_2SO_4), C-Na_2SO_4
gel-6	6	/	/	1.14	thenardite (Na_2SO_4), C-Na_2SO_4
gel-8	8	/	/	1.24	thenardite (Na_2SO_4)
gel-10	10	/	/	0.97	C-Na_2SO_4 (*Cmcm*(63))
gel-12	12	/	/	1.02	thenardite (Na_2SO_4), S-sodalite
gel-14	14	/	/	1.10	thenardite (Na_2SO_4), S-sodalite
SOD-2-4	4	100	48	0.13	C-Na_2SO_4, natroalunite
SOD-2-6	6	100	48	0.49	C-Na_2SO_4 (*Cmcm*(63))
SOD-2-8	8	100	48	1.14	thenardite (Na_2SO_4)
SOD-2-10	10	100	48	1.06	thenardite (Na_2SO_4)
SOD-2-12	12	100	48	1.11	S-sodalite, thenardite (Na_2SO_4)
SOD-2-14	14	100	48	1.11	S-sodalite

* There are two polymorphs of Na_2SO_4 observed in the system. One is thenardite (Na_2SO_4, *Fddd*(70), PDF#37-1465) while the other is C-Na_2SO_4 (*Cmcm*(63), PDF#79-1553).

Table S2 Powder X-ray diffraction pattern indexing and refinement for SOD-1 (B-sodalite)

(*h k l*)	2θ (Calc.)	2θ (Obs.)	Δ2θ	*d* (Calc.)	*d* (Obs.)	*I* (height)%.	*I* (Area)%
(1 1 0)	14.061	14.041	0.020	6.2932	6.3022	47	39
(2 0 0)	19.936	19.944	-0.008	4.4500	4.4483	4	4
(2 1 0)	22.318	22.310	0.008	3.9802	3.9815	1.5	2
(2 1 1)	24.479	24.472	0.007	3.6334	3.6344	100	100
(2 2 0)	28.340	28.344	-0.005	3.1466	3.1461	3	3
(2 2 1)	30.098	30.072	0.026	2.9666	2.9691	1.6	3
(0 1 3)	31.768	31.768	0.000	2.8144	2.8145	24	29
(2 2 2)	34.893	34.888	0.005	2.5692	2.5696	30	37
(1 2 3)	37.790	37.784	0.006	2.3786	2.3790	7	9
(4 1 1)	43.086	43.080	0.006	2.0977	2.0980	23	33
(0 2 4)	45.543	45.561	-0.017	1.9901	1.9894	1.1	1.4
(3 3 2)	47.901	47.929	-0.028	1.8975	1.8964	0.9	1.4
(4 2 2)	50.175	50.183	-0.008	1.8167	1.8164	4	7
(1 3 4)	52.376	52.376	0.000	1.7454	1.7454	6	10
(5 2 1)	56.595	56.600	-0.006	1.6249	1.6248	0.5	0.7
(4 4 0)	58.628	58.632	-0.004	1.5733	1.5732	5	10

* Refined cell parameters: $a = b = c = 8.8999$ (3) Å, $\alpha = \beta = \gamma = 90^\circ$, $V = 704.96$ Å^3, space group: $P\bar{4}3n$, $|\Delta 2\theta| = 0.0053^\circ$, F(12) = 126.7(18). Abbreviation: Calc. and Obs. are abbreviated for calculated and observed, respectively.

Table S3 Powder X-ray diffraction pattern indexing and refinement for SOD-2 using a cubic cell

(*h k l*)	2θ (Calc.)	2θ (Obs.)	Δ2θ	*d* (Calc.)	*d* (Obs.)	*I* (Obs.)%.	*I* (Calc.)*	Note
(1 1 0)	13.856	13.880	-0.025	6.3861	6.3748	73.6	34.7	
	22.281				3.9866	11.0		Diaspore
(2 1 1)	24.118	24.168	-0.050	3.6870	3.6795	100.0	100.0	
(0 1 3)	31.294	31.302	-0.008	2.8560	2.8552	9.1	13.9	
(2 2 2)	34.369	34.312	0.058	2.6071	2.6114	26.8	25.5	
(3 3 0)	42.428	42.456	-0.027	2.1287	2.1274	20.1	8.3	
(5 1 0)	51.557	51.608	-0.051	1.7712	1.7696	9.8	14.0	
(4 4 0)	57.694	57.643	0.051	1.5965	1.5978	4.7	9.6	
	57.911				1.5899	7.1		Unindexed

* Refined cell parameters: $a = 9.031(4)$ Å, $V = 736.64$ Å^3, space group: $P\bar{4}3n$, $|\Delta 2\theta| = 0.0386^\circ$, F(7) = 25.9(7). The calculated intensities were obtained from the calculation of using single crystal data of nosean (I. Hassan, H.D. Grundy, Can. Mineral. 27 (1989) 165-172). Abbreviation: Calc. and Obs. are abbreviated for calculated and observed, respectively.

Table S4 Powder X-ray diffraction pattern indexing and refinement for SOD-2 using a rhombohedral cell

(*h k l*)	2θ (Calc.)	2θ (Obs.)	Δ2θ	*d* (Calc.)	*d* (Obs.)	*I* (height)%.	*I* (Area)%	CS(Å)
(1 0 1)	13.904	13.880	0.023	6.3642	6.3748	73	49	627
(2 1 1)	24.144	24.168	-0.023	3.6830	3.6795	100	100	368
(1 3 1)	31.314	31.289	0.024	2.8542	2.8564	9	13	
(4 0 1)	34.387	34.311	0.076	2.6058	2.6114	28	37	260
(4 1 0)	37.216	37.192	0.025	2.4140	2.4155	5	5	
(0 5 1)	42.442	42.456	-0.014	2.1281	2.1274	20	28	253
(1 5 2)	51.612	51.608	0.004	1.7694	1.7696	9	12	284
(4 4 0)	57.688	57.687	0.001	1.5967	1.5967	7	8	
(4 0 4)	57.911	57.912	-0.001	1.5910	1.5910	7	10	

* CS denotes crystallite size. Refined cell parameters: trigonal system, $a = b = 12.773$ (3), $c = 7.781$ (3) Å, $\alpha = \beta = 90$, $\gamma = 120^{\circ}$, $V = 1099.43$ Å^3, space group: $R3m$. Abbreviation: Calc. and Obs. are abbreviated for calculated and observed, respectively.

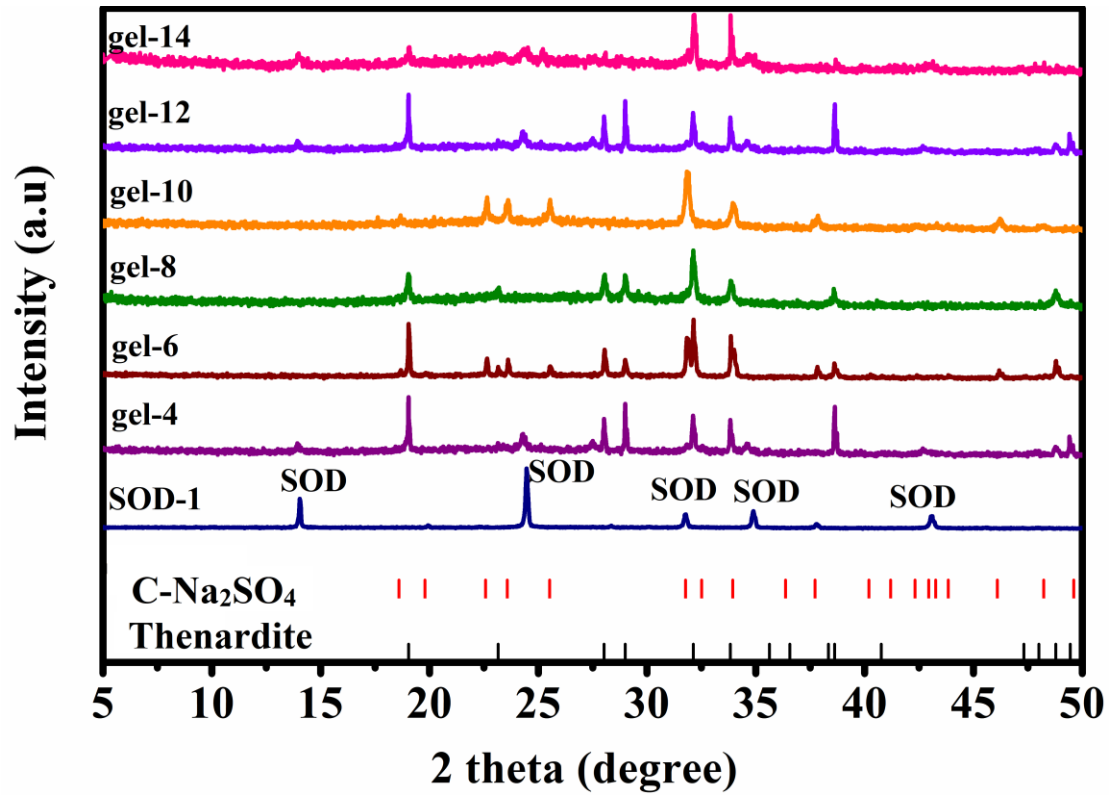

Figure S1 PXRD patterns of the dried gels formed at different pH values

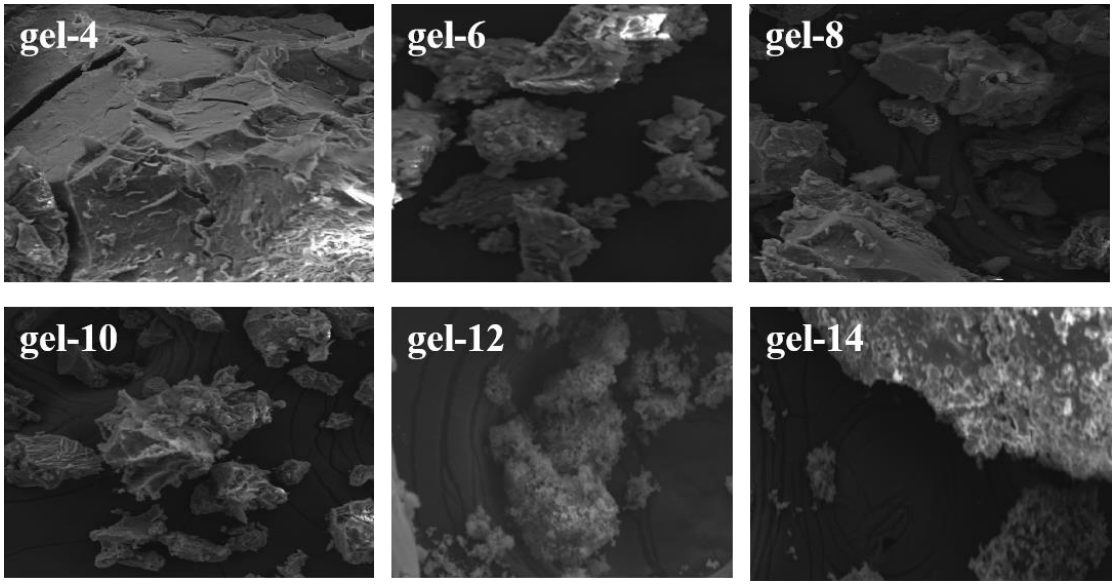

Figure S2 SEM images of the dried gels formed at different pH values

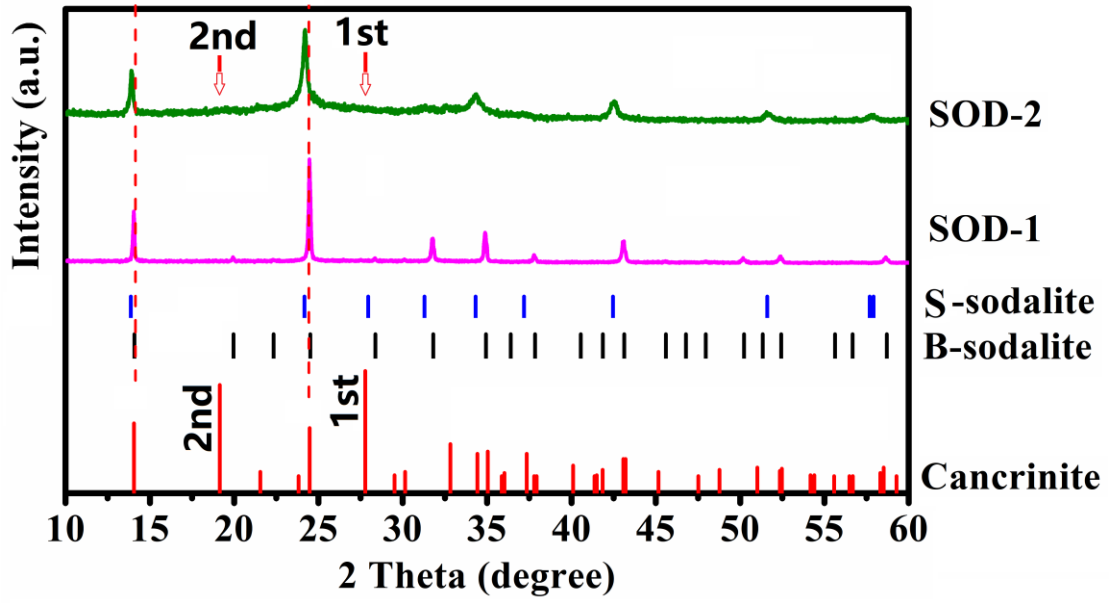

Figure S3 Comparison of PXRD patterns of SOD-1 and SOD-2, and Bragg positions among B-Sodalite, S-sodalite and cancrinite (PDF #34-0176). The arrows labelled 1st and 2nd in SOD-2 PXRD pattern indicate the positions of cancrinite.

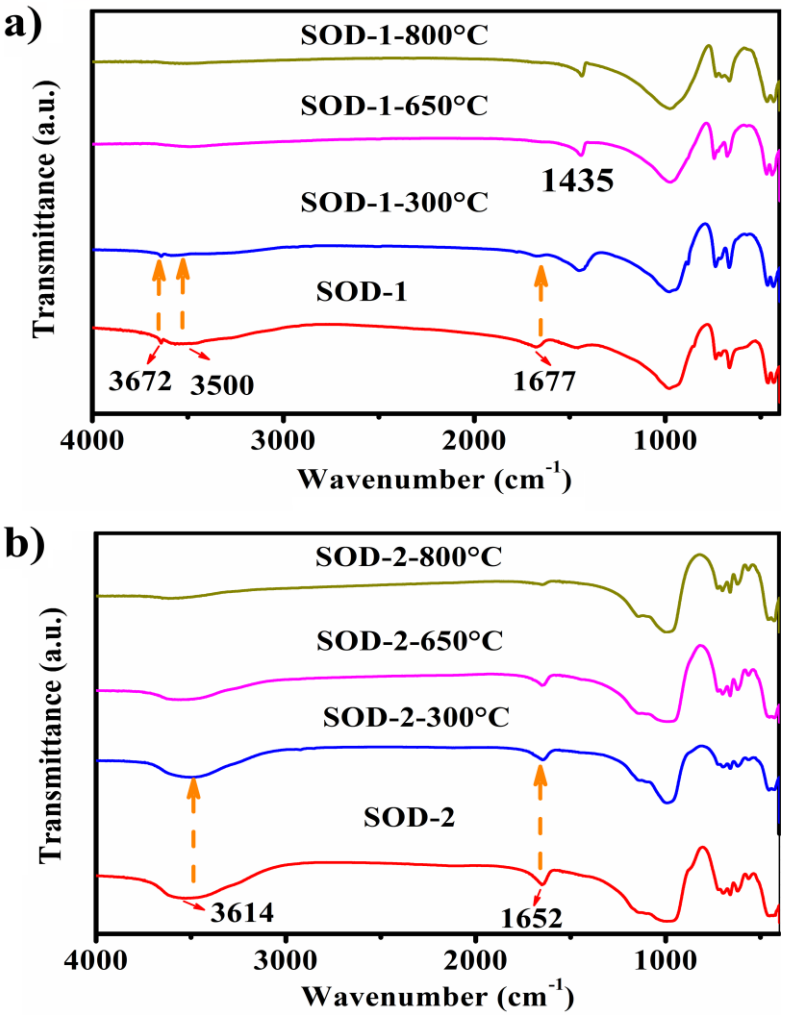

Figure S4 Comparison of *ex-situ* FT-IR spectra of SOD-1 and SOD-2 samples (the as-is and those after calcination at 300, 650 and 800 °C).

Table S5 The chemical composition of samples after calcination at 800 °C measured from a CHNOS elemental analyzer

Elements	C (wt.%)	H (wt.%)	S (wt.%)
SOD-1	0.7330	0.1216	0.1478
SOD-1	1.0047	0.1710	0.1932
SOD-2	0.3927	0.5584	2.1038
SOD-2	0.4245	0.5879	2.9233

Note that the samples after calcination at 800 °C for 0.5 h still contain water molecules, which could be absorbed from the air during the waiting time before analyses. The analytical laboratory is located on an island surrounded by the sea.

2.

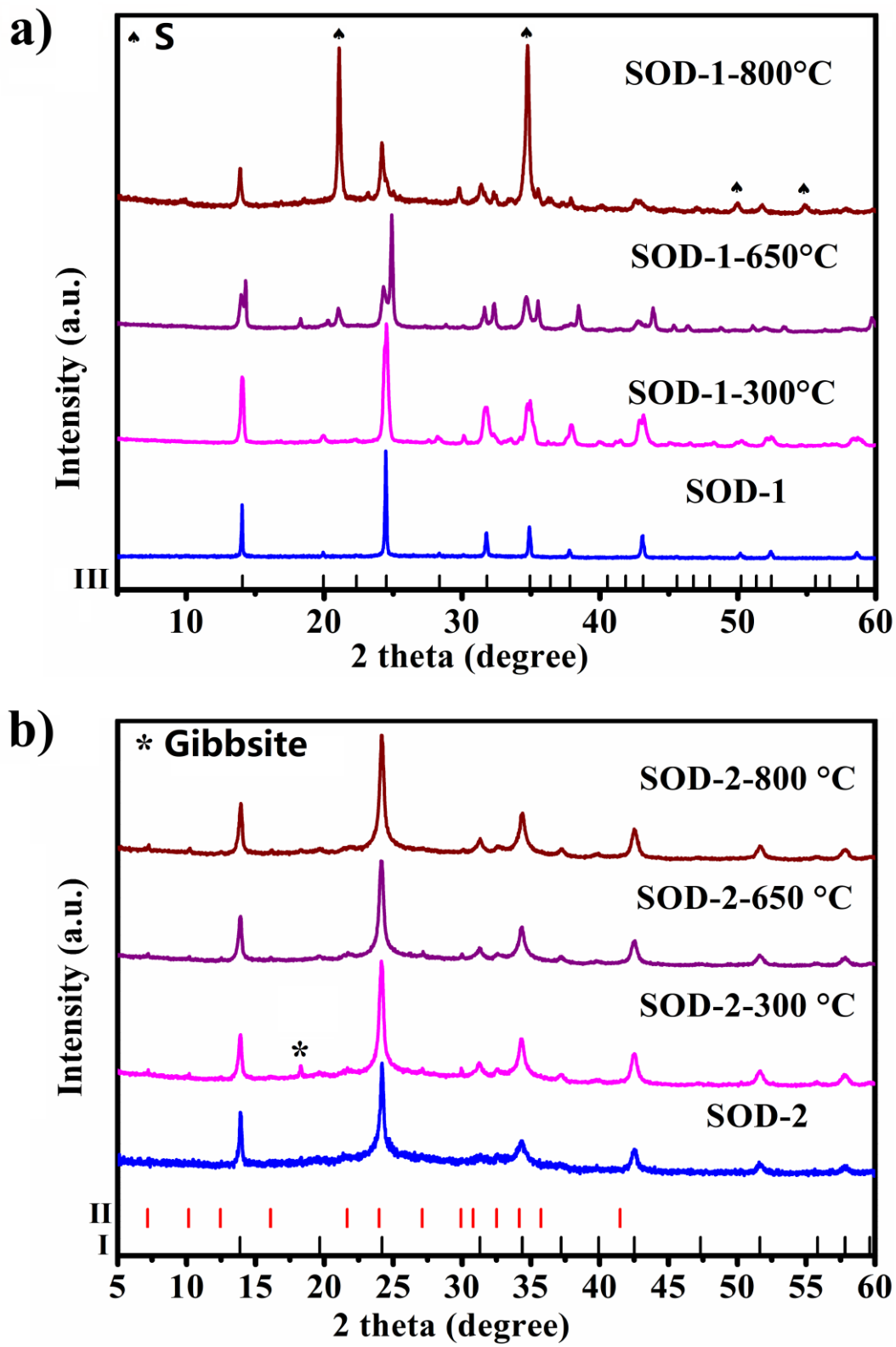

Figure S5 Comparison of *ex-situ* PXRD patterns of the samples of SOD-1 (a) and SOD-2 (b) calcined at 300 ˚C, 650 ˚C, and 800 ˚C, respectively. The vertical bars below the patterns for I, II, and III are the Bragg positions of Zeolite A, S-sodalite, and B-sodalite, respectively. The capitalized letter of S indicates the presence of a sodium aluminum silicate (PDF#10-0033).

Table S6 Chemical composition of samples from EDX analysis

Samples	Element	Weight%	Atomic%
SOD-1	O K	55.74	67.22
	Na K	13.29	11.15
	Al K	14.46	10.34
	Si K	15.94	10.95
	S K	0.46	0.28
	Ca K	0.12	0.06
SOD-2	O K	52.33	64.3
	Na K	14.05	12.02
	Al K	14.42	10.51
	Si K	16.70	11.69
	S K	2.12	1.30
	Ca K	0.39	0.19

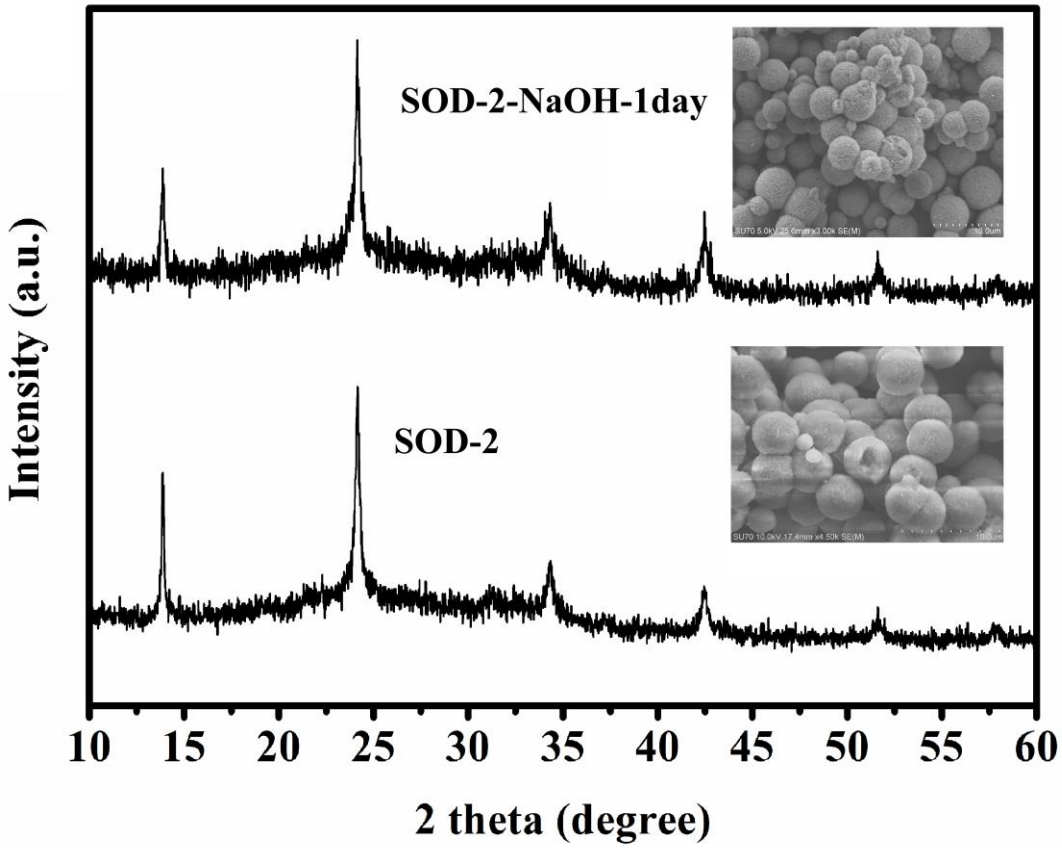

Figure S6 Comparison of PXRD patterns and SEM images between the pristine sample and the sample immersed in NaOH for 24 h

Alkali-resistant test of S-sodalite: The sample was immersed in NaOH solution with pH > 14 for 24 h and the recouped powder was dried overnight in an oven at 120° C prior to be X-rayed.