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Supplementary information

3D reconstruction of atomic structures from high angle annular dark field (HAADF) STEM images and its application on zeolite silicalite-1

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Table S1 Plane group determination from the HAADF-STEM and HRTEM images, respectively for the three main zone axes. R_A% is the R-value of the amplitudes, ϕ_{Res} corresponds to the average phase errors in degrees and A_o/A_e the ratio between of the amplitude sum of the odd reflections that should have been absent by the symmetry and the amplitude sum of the corresponding even reflections.

	HAADF-STEM				HRTEM			
	Plane group	R_A%	ϕ_{Res}	A_o/A_e	Plane group	R_A%	ϕ_{Res}	A_o/A_e
[010]	p1	–	–	–	p1	–	–	–
	p2	–	6.0	–	p2	–	22.7	–
	pm m|x	4.3	16.8	–	pm m|x	11.6	17.8	–
	pm m|y	4.3	16.5	–	pm m|y	11.6	22.2	–
	pg g|x	4.3	3.2	0.2	pg g|x	11.7	9.7	0.1
	pg g|y	4.3	3.9	–	pg g|y	11.8	19.2	0.3
	cm m|x	3.1	2.8	0.4	cm m|x	10.9	8.9	0.5
	cm m|y	3.1	2.8	0.4	cm m|y	10.9	17.2	0.5
	pmm	4.3	19.2	–	pmm	11.6	29.9	–
	pmg m|x	4.3	19.8	–	pmg m|x	11.8	29.7	0.3
	pmg m|y	4.3	19.1	0.2	pmg m|y	11.7	27.1	0.1
	pgg	4.3	6.0	0.2	pgg	11.8	24.0	0.3
	cmm	3.1	4.7	0.4	cmm	10.9	21.4	0.5
[001]	p1	–	–	–	p1	–	–	–
	p2	–	9.9	–	p2	–	23.6	–
	pm m|x	8.1	5.0	–	pm m|x	8.5	15.5	–
	pm m|y	8.1	23.9	–	pm m|y	8.5	22.0	–
	pg g|x	10.1	19.6	3.5	pg g|x	9.8	27.6	3.0
	pg g|y	8.2	6.7	0.5	pg g|y	8.6	11.7	0.4
	cm m|x	6.1	5.2	0.8	cm m|x	7.2	19.0	1.1
	cm m|y	6.1	5.2	0.8	cm m|y	7.2	6.1	1.1
	pmm	8.1	28.5	–	pmm	8.5	40.1	–
	pmg m|x	8.2	9.7	0.5	pmg m|x	8.6	24.2	0.4
	pmg m|y	10.1	23.0	3.5	pmg m|y	9.8	37.7	3.0
	pgg	10.2	28.2	1.7	pgg	9.9	37.1	2.1
	cmm	6.1	8.5	0.8	cmm	7.2	22.2	1.1
[100]	p1	–	–	–	p1	–	–	–
	p2	–	20.9	–	p2	–	26.0	–
	pm m|x	6.6	13.2	–	pm m|x	17.7	14.0	–
	pm m|y	6.6	9.4	–	pm m|y	17.7	17.3	–
	pg g|x	6.6	13.6	0.7	pg g|x	17.8	14.2	0.2
	pg g|y	6.6	9.6	0.1	pg g|y	17.8	17.7	0.0
	cm m|x	5.6	13.1	0.3	cm m|x	17.5	13.8	0.2
	cm m|y	5.6	9.2	0.3	cm m|y	17.5	17.2	0.2
	pmm	6.6	21.6	–	pmm	17.7	28.0	–
	pmg m|x	6.6	21.1	0.1	pmg m|x	17.8	29.0	0.0
	pmg m|y	6.6	21.8	0.7	pmg m|y	17.8	28.3	0.2
	pgg	6.6	21.6	0.3	pgg	17.8	28.2	0.2
	cmm	5.6	20.5	0.3	cmm	17.5	27.7	0.2

Table S2 Comparison of crystallographic structure factors calculated from the structure model of silicalite-1 (MFI) with the amplitudes and phases extracted from the HAADF-STEM and HRTEM images acquired along the [010] projection. Erroneous experimental phases are marked in red. The amplitudes are scaled based on the strongest amplitude and the phases extracted from the HRTEM images are shifted by 180° in order to compensate for the reversed contrast in the HRTEM images.

				Calculated			HAADF-STEM			HRTEM	
h	k	l	d-value	Amp	Phase		Amp	Phase		Amp	Phase
1	0	1	11.00	5189	0		9796	0		10000	0
2	0	0	10.05	6615	180		10000	180		9894	180
2	0	1	7.98	1279	180		585	180		518	180
0	0	2	6.57	5189	180		3972	180		9091	180
1	0	2	6.25	2585	180		3702	180		4566	180
3	0	1	5.97	3566	0		4333	0		7965	0
2	0	2	5.50	2768	180		3082	180		6805	180
4	0	0	5.02	3567	180		3021	180		6541	180
4	0	1	4.69	1398	180		-	-		310	0
1	0	3	4.28	2081	180		979	180		1481	180
2	0	3	4.02	3509	180		935	180		1119	180
4	0	2	3.99	563	180		232	180		1527	180
5	0	1	3.84	10000	180		5761	180		6834	180
3	0	3	3.67	7390	180		4442	180		4872	180
6	0	0	3.35	4705	180		1833	180		2205	180
4	0	3	3.30	539	0		1003	180		876	0
0	0	4	3.29	3037	0		1112	0		2317	0
6	0	1	3.24	4940	0		639	0		485	0
1	0	4	3.24	3960	180		1726	180		1212	180
2	0	4	3.12	627	180		743	180		750	180
5	0	3	2.96	4736	0		1805	0		1048	0
3	0	4	2.95	734	0		-	-		267	0
7	0	1	2.80	638	0		329	0		476	0
4	0	4	2.75	504	180		480	0		323	0
6	0	3	2.66	1991	0		287	180		449	180
7	0	2	2.63	3256	0		203	180		259	180
1	0	5	2.61	2373	0		434	0		412	180
8	0	0	2.51	2997	180		1030	180		812	180
3	0	5	2.45	3123	0		755	0		466	180
7	0	3	2.40	1639	0		950	0		490	180
8	0	2	2.35	1316	0		1090	0		605	0
4	0	5	2.33	304	180		303	180		-	-
9	0	1	2.20	1054	0		-	-		232	180
5	0	5	2.20	1046	180		1104	180		-	-
0	0	6	2.19	3421	0		955	0		-	-
1	0	6	2.18	495	180		373	180		-	-
9	0	2	2.11	986	0		-	-		294	180
6	0	5	2.07	1217	0		335	180		-	-
10	0	0	2.01	5597	0		304	0		568	180
4	0	6	2.01	196	0		239	180		-	-
8	0	4	2.00	1697	0		325	0		343	0
6	0	6	1.83	67	180		296	180		-	-

Table S3 Comparison of crystallographic structure factors calculated from the structure model of silicalite-1 (MFI) with the amplitudes and phases extracted from the HAADF-STEM and HRTEM images acquired along the [001] projection. Erroneous experimental phases are marked in red. The amplitudes are scaled based on the strongest amplitude and the phases extracted from the HRTEM images are shifted by 180° in order to compensate for the reversed contrast in the HRTEM images.

				Calculated			HAADF-STEM			HRTEM	
h	k	l	d-value	Amp	Phase		Amp	Phase		Amp	Phase
2	0	0	10.05	9101	180		8796	180		4904	180
0	2	0	9.87	10000	0		10000	0		10000	0
2	1	0	8.95	1340	0		1126	0		846	0
2	2	0	7.04	895	180		468	180		1080	0
2	3	0	5.50	766	180		1273	180		2420	180
4	0	0	5.02	4907	180		2695	180		4463	180
0	4	0	4.93	7209	180		3317	180		6209	180
4	1	0	4.87	1743	0		925	0		2175	0
4	2	0	4.48	1592	180		414	180		421	180
2	4	0	4.43	778	0		375	0		783	0
4	3	0	3.99	4519	0		1085	0		1807	0
2	5	0	3.67	2259	180		673	180		700	180
4	4	0	3.52	593	180		229	0		223	0
6	0	0	3.35	6473	180		1878	180		3082	180
6	1	0	3.30	756	180		317	180		310	180
0	6	0	3.29	4368	0		1841	0		2219	0
6	2	0	3.17	4	0		303	180		-	-
2	6	0	3.13	1760	180		-	-		214	180
4	5	0	3.10	1619	180		545	180		492	180
6	3	0	2.98	1378	0		773	0		897	0
6	5	0	2.55	1437	0		367	0		345	0
8	0	0	2.51	4123	180		1807	180		698	180
8	1	0	2.49	1451	180		-	-		223	0
0	8	0	2.47	3532	180		1923	180		819	180
4	7	0	2.46	5743	180		378	180		467	180
8	2	0	2.43	2053	180		470	180		-	-
2	8	0	2.40	1230	0		407	0		308	0
10	0	0	2.01	7701	0		455	0		359	180
0	10	0	1.97	9633	180		441	180		615	180

Table S4 Comparison of crystallographic structure factors calculated from the structure model of silicalite-1 (MFI) with the amplitudes and phases extracted from the HAADF-STEM and HRTEM images acquired along the [100] projection. Erroneous experimental phases are marked in red. The amplitudes are scaled based on the strongest amplitude and the phases extracted from the HRTEM images are shifted by 180° in order to compensate for the reversed contrast in the HRTEM images.

				Calculated			HAADF-STEM			HRTEM	
h	k	l	d-value	Amp	Phase		Amp	Phase		Amp	Phase
0	1	1	10.94	6575	180		7499	180		6825	180
0	2	0	9.87	10000	0		10000	0		10000	0
0	0	2	6.57	7139	180		2953	180		6853	180
0	3	1	5.88	4221	0		3080	0		5674	0
0	2	2	5.47	3116	180		2033	180		3999	180
0	4	0	4.93	7209	180		3337	180		6415	180
0	1	3	4.28	2525	0		1177	0		3740	0
0	4	2	3.95	3383	0		-	-		384	180
0	5	1	3.78	11700	0		4896	0		6217	0
0	3	3	3.65	6326	180		1554	180		2584	180
0	6	0	3.29	4368	0		1742	0		1626	0
0	0	4	3.29	4178	0		434	0		582	180
0	2	4	3.12	2016	180		278	180		737	180
0	6	2	2.94	2171	0		-	-		369	0
0	5	3	2.93	6417	180		1326	180		1894	180
0	7	1	2.76	147	0		252	0		472	0
0	4	4	2.73	1310	180		-	-		369	180
0	1	5	2.61	116	0		-	-		731	0
0	8	0	2.47	3532	180		1235	180		506	180
0	3	5	2.44	3584	180		679	180		618	180
0	6	4	2.32	458	0		-	-		309	180
0	8	2	2.31	949	0		336	180		549	180
0	0	6	2.19	4707	0		598	0		303	0
0	5	5	2.19	748	180		586	180		435	180
0	2	6	2.14	1219	0		268	0		-	-
0	10	0	1.97	9633	180		335	180		778	180
0	8	4	1.97	6520	180		304	180		263	0
0	3	7	1.81	173	180		205	180		215	0