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Supporting Information

Impact of chabazite SSZ-13 textural properties and chemical composition on CO_2 adsorption applications

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X-ray diffraction

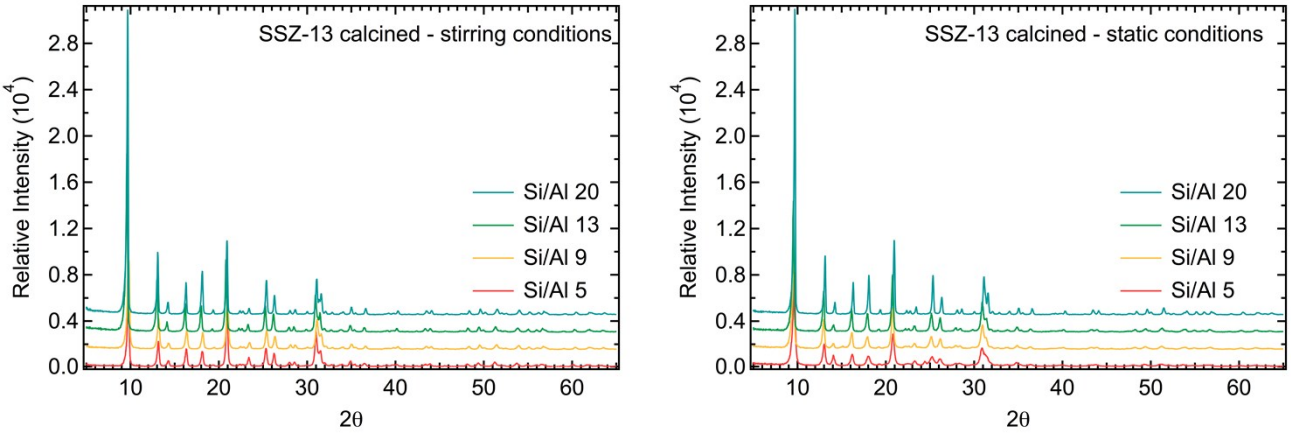


Figure S1. PXRD patterns of materials obtained under stirring (left) and static (right) conditions after calcination. Color-coding is reported in the legend.

Rietveld refinement

Table S1. Lattice parameters determined for the PXRD data using Rietveld Refinement with TOPAS software. All fits were corrected for the sample displacement and zero shift.

Sample	a (Å)	c (Å)	V (Å³)	Sample displacement (mm)
Static				
Si/Al 5 as-made	13.648(1)	14.996(1)	2419.19(5)	-0.040(1)
Si/Al 5 calcined	13.692(2)	14.857(2)	2412.1(7)	-0.194(3)
Si/Al 9 as-made	13.589(1)	14.945(1)	2390.0(2)	-0.065(1)
Si/Al 9 calcined	13.610(1)	14.822(2)	2377.7(5)	0.004(2)
Si/Al 13 as-made	13.564(1)	14.947(1)	2381.4(1)	-0.091(1)
Si/Al 13 calcined	13.593(1)	14.797(1)	2367.9(4)	-0.019(1)
Si/Al 20 as-made	13.537(1)	14.938(1)	2370.8(3)	-0.002(1)
Si/Al 20 calcined	13.582(1)	14.786(1)	2362.2(2)	-0.162(1)
Stirring				
Si/Al 5 as-made	13.666(1)	15.015(1)	2428.6(3)	-0.202(2)
Si/Al 5 calcined	13.691(1)	14.842(1)	2409.3(1)	-0.182(1)
Si/Al 9 as-made	13.610(1)	14.969(1)	2401.4(1)	-0.079(1)
Si/Al 9 calcined	13.637(1)	14.815(1)	2386.1(3)	-0.340(1)
Si/Al 13 as-made	13.576(1)	14.949(1)	2386.2(2)	-0.198(1)
Si/Al 13 calcined	13.608(1)	14.778(1)	2369.9(2)	-0.245(1)
Si/Al 20 as-made	13.533(1)	14.928(1)	2367.6(1)	-0.110(1)
Si/Al 20 calcined	13.584(1)	14.736(1)	2354.9(2)	-0.126(1)

Helium ion microscopy

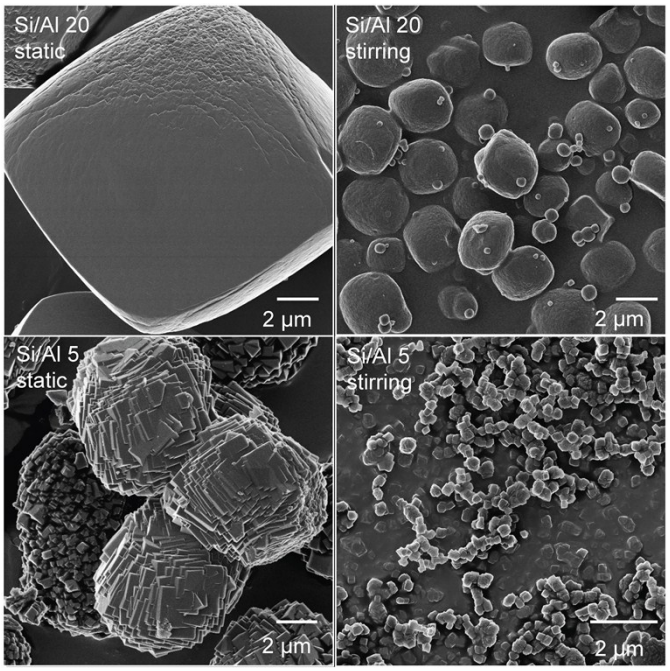


Figure S2. HIM images comparing Al-poor and Al-rich material obtained in stirring and static conditions respectively. The magnification is reported in the legend.

Particle size distribution

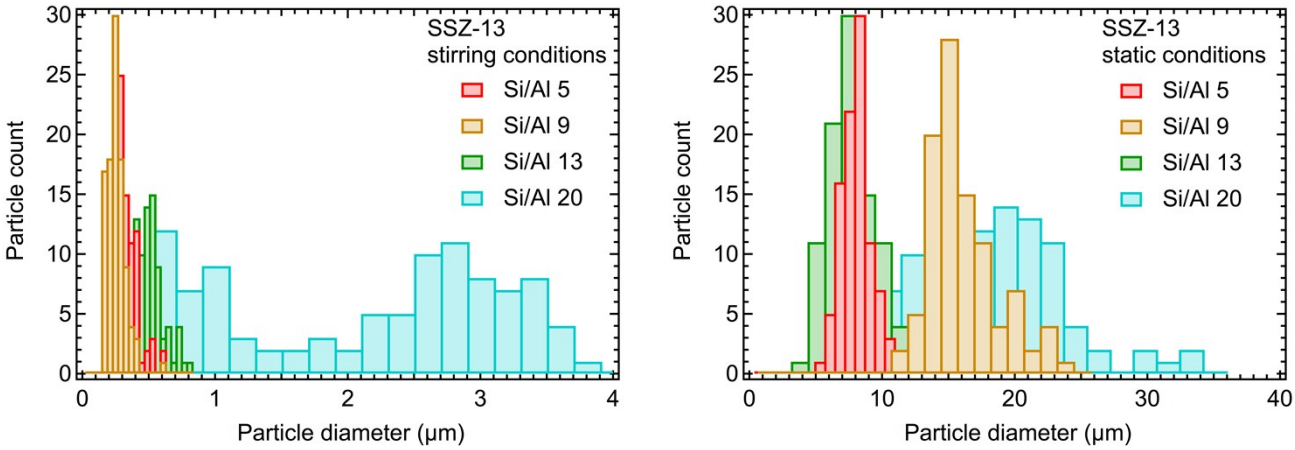


Figure S3. Particle size distribution reflecting the mean particle size of synthesized samples. On average the diameter of 100 particles was measured and used to determine the mean.

^{27}Al MAS NMR

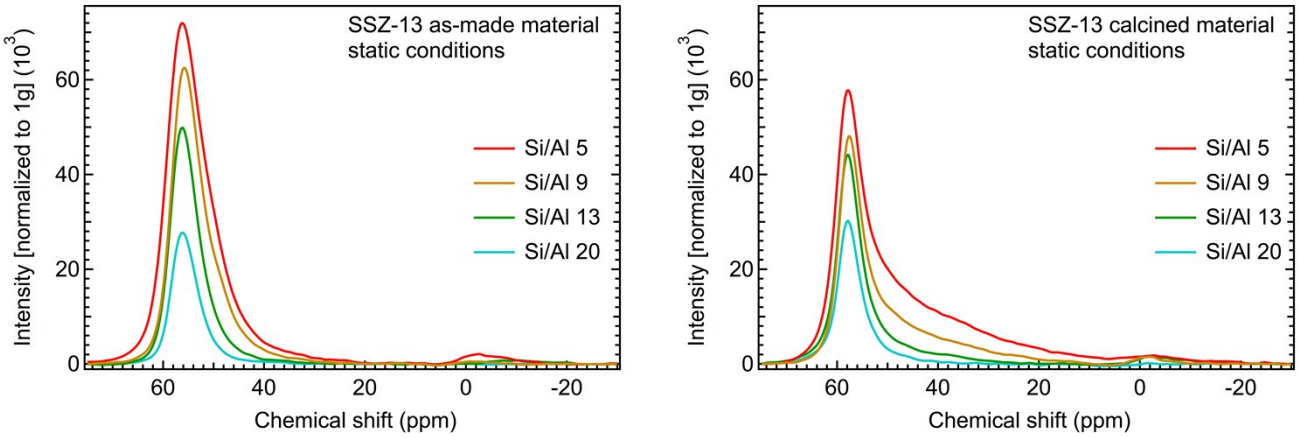


Figure S4. ^{27}Al MAS NMR spectra taken for SSZ-13 obtained in static conditions before (left) and after calcination (right). Note the significant degree of penta-coordinated aluminium being formed as a result of calcination. Color-coding is reported in the legend.

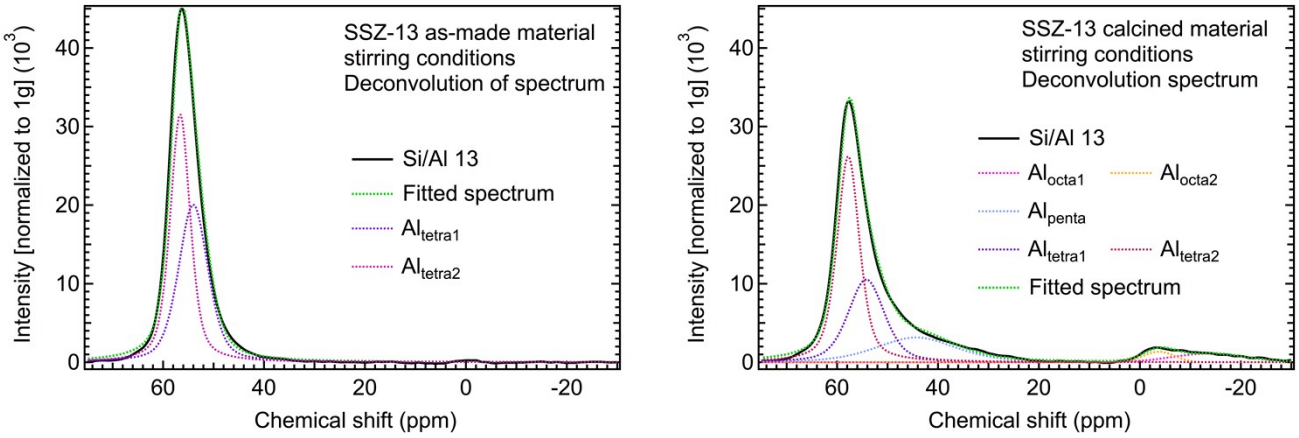


Figure S5. Example of deconvolution of ^{27}Al MAS NMR spectra used to determine contribution of different Al coordination spheres. Gaussian line shapes were used, requiring the use of two peaks for the tetrahedral region. The line shape was kept constant for all peaks of a specific Al coordination, allowing the width, chemical shift and peak height to optimize itself to the least number of squares. The consistent manner of this procedure allowed us to make the statements regarding the contribution of specific Al species within adequate measure.

N_2-sorption

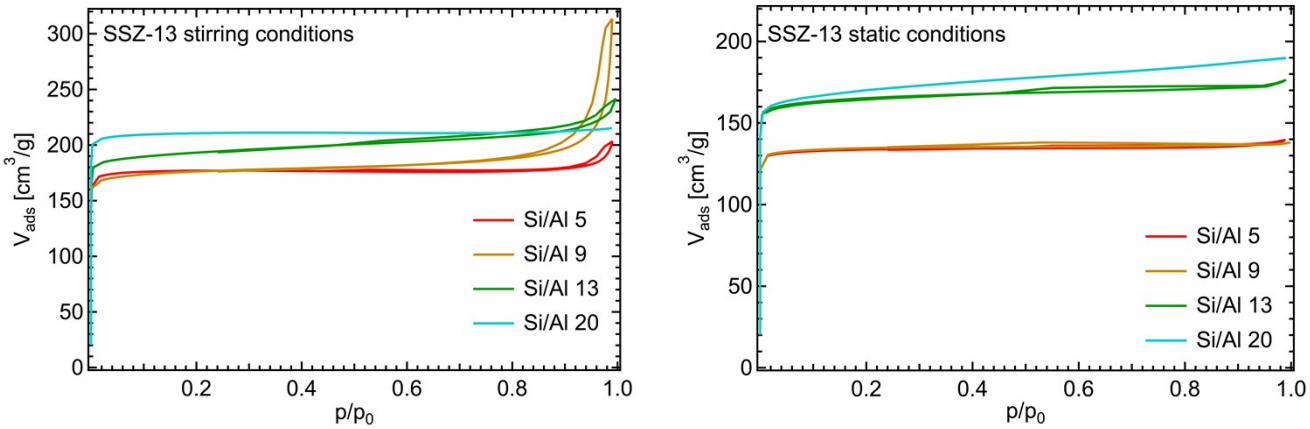


Figure S6. N_2-physisorption isotherms for both SSZ-13 synthesized in stirring and static conditions. Color-coding is reported in the legend.

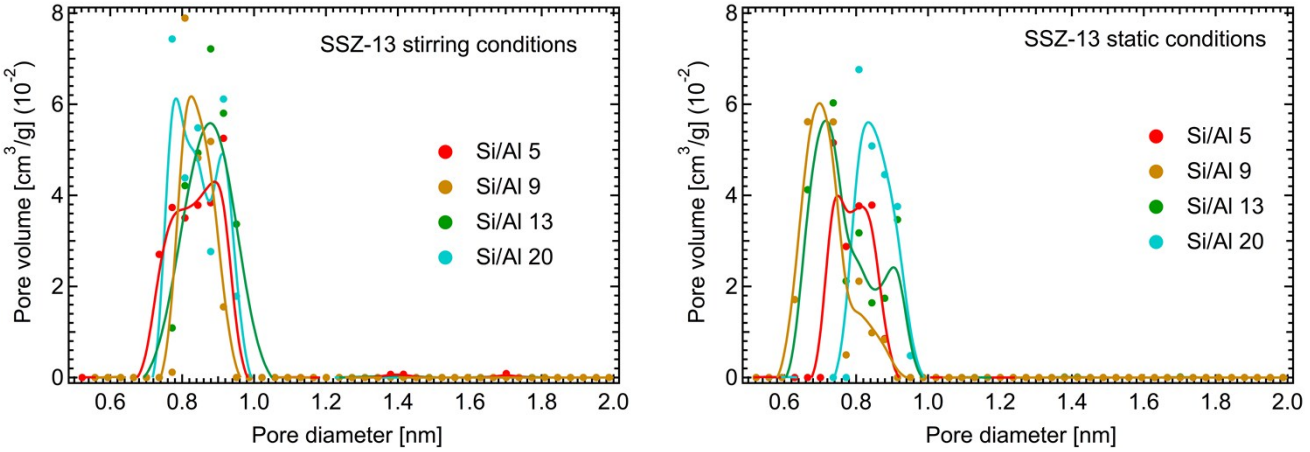


Figure S7. Pore size distribution using the pore volume determined with the NL-DFT method. The CHA cage volume is typically cited to be between 0.7 and 1.2 nm. A spline fit was used for better visualization. Narrower pore size distribution is observed for material synthesized in stirring conditions. No noticeable mesopore contribution up to 380 nm could be determined.

CO_2 and N_2 adsorption measurements

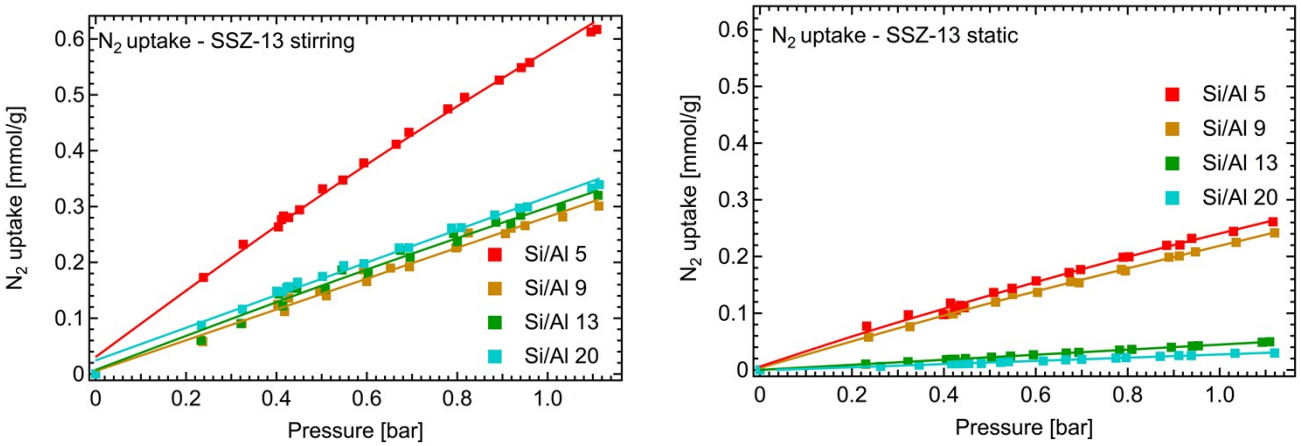
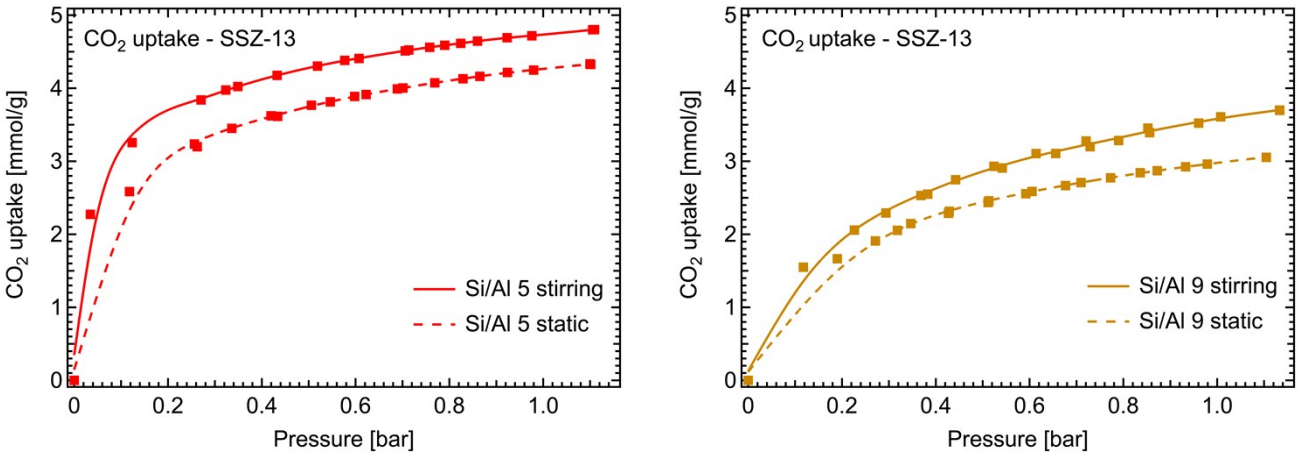


Figure S8. N_2 adsorption measurements at N_2 pressures up to 1 bar. Only negligible amounts of N_2 are adsorbed on SSZ-13 synthesized in either stirring (left) or static (right) conditions. Color-coding is reported in the legend.



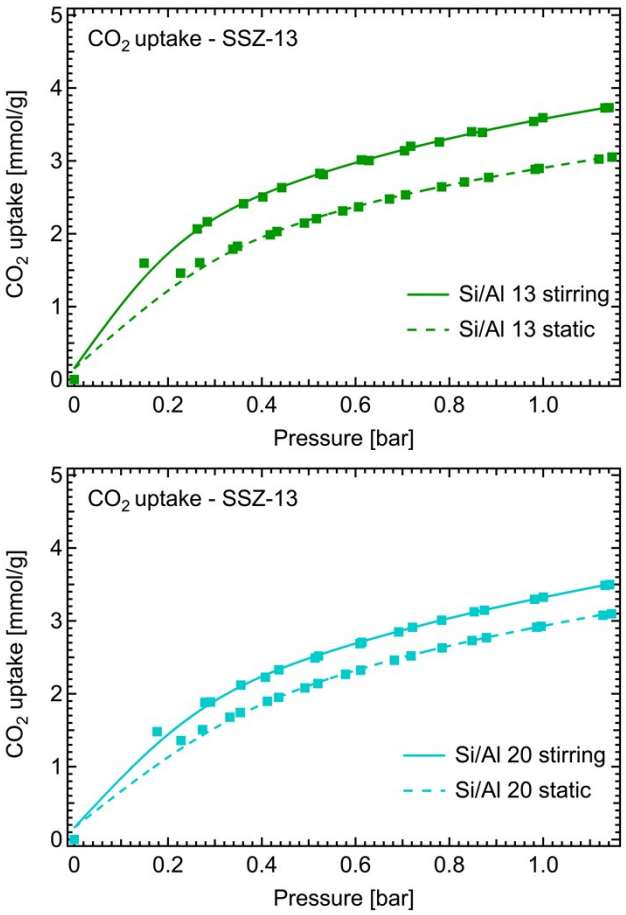


Figure S9. CO_2 adsorption measurements at CO_2 pressures up to 1 bar. A comparison between material obtained in stirring and static conditions is made for several different Si/Al ratios. All SSZ-13 synthesized with stirring conditions exhibits superior CO_2 adsorption capabilities. The color-coding is reported in the legend.

Table S2. Crystallite sizes, and agreement indices from the Rietveld refinements

Sample ID	Crystallite size (nm)	GOF	R_Bragg	Rexp	wRp	Rp
Si/Al 5 static as-made	56.7(8)	1.60	0.978	7.66	12.24	9.06
Si/Al 5 static calcined	41.2(5)	1.93	1.555	7.07	13.63	10.60
Si/Al 5 stirring as-made	62.5(6)	1.40	0.821	7.61	10.69	7.89
Si/Al 5 stirring calcined	69.6(8)	1.54	1.070	7.37	11.36	8.87
Si/Al 9 static as-made	67.7(9)	1.59	1.017	7.57	12.06	9.28
Si/Al 9 static calcined	60.3(8)	1.77	1.270	7.27	12.86	9.98
Si/Al 9 stirring as-made	71.7(7)	1.31	0.624	7.47	9.76	7.09
Si/Al 9 stirring calcined	60.5(6)	1.45	0.897	7.38	10.73	8.38

Si/Al 13 static as-made	106(2)	1.57	0.999	7.47	11.71	8.79
Si/Al 13 static calcined	113(2)	1.79	1.264	7.06	12.65	9.74
Si/Al 13stirring as-made	90(1)	1.40	0.689	7.45	10.40	7.40
Si/Al 13stirring calcined	112(1)	1.53	0.834	6.85	10.49	8.14
Si/Al 20 static as-made	203(4)	1.58	0.946	7.62	12.05	8.66
Si/Al 20 static calcined	340(10)	1.91	1.321	6.69	12.81	9.91
Si/Al 20 stirring as-made	211(4)	1.50	0.765	7.38	11.05	7.76
Si/Al 20 stirring calcined	260(6)	1.87	1.189	6.56	12.28	9.47

Crystallite size is *LVol-FWHM* from TOPAS; fit criteria are goodness of fit as chi^2 (GOF), R_Bragg, R-expected (Rexp), R-weighted pattern (wRp), and R-pattern (Rp) from TOPAS. While the GOF values (chi-squared < 2) suggest good refinements, due to the quality of the present XRD data we cannot expect the R Bragg, Rwp and Rp fit criteria to be as low for high-quality synchrotron data (often Rwp < 0.15 and R Bragg < 0.20 may be achieved when atomic coordinates and site occupancies are refined for perfectly polycrystalline phases from synchrotron data). The present data is lower-quality desktop diffractometer data, which is appropriate for extracting lattice parameters and crystallite sizes, but due to lower instrumental resolution and signal-to-noise ratio, it will not result in very low Rwp and R Bragg values. The parameter Rexp our refinement program TOPAS gives (also included in the Table) is an expected value for the goodness of fit; it reflects the "quality" of our diffraction pattern and gives us an idea of the theoretical minimum Rwp that can be reached. The actual Rwp obtained is usually greater than this Rexp value, but the general rule-of-thumb in the crystallographer community is that if the Rwp is close to 2*Rexp, then the fit is acceptable. Our refinement agreement indices meet this criterion.

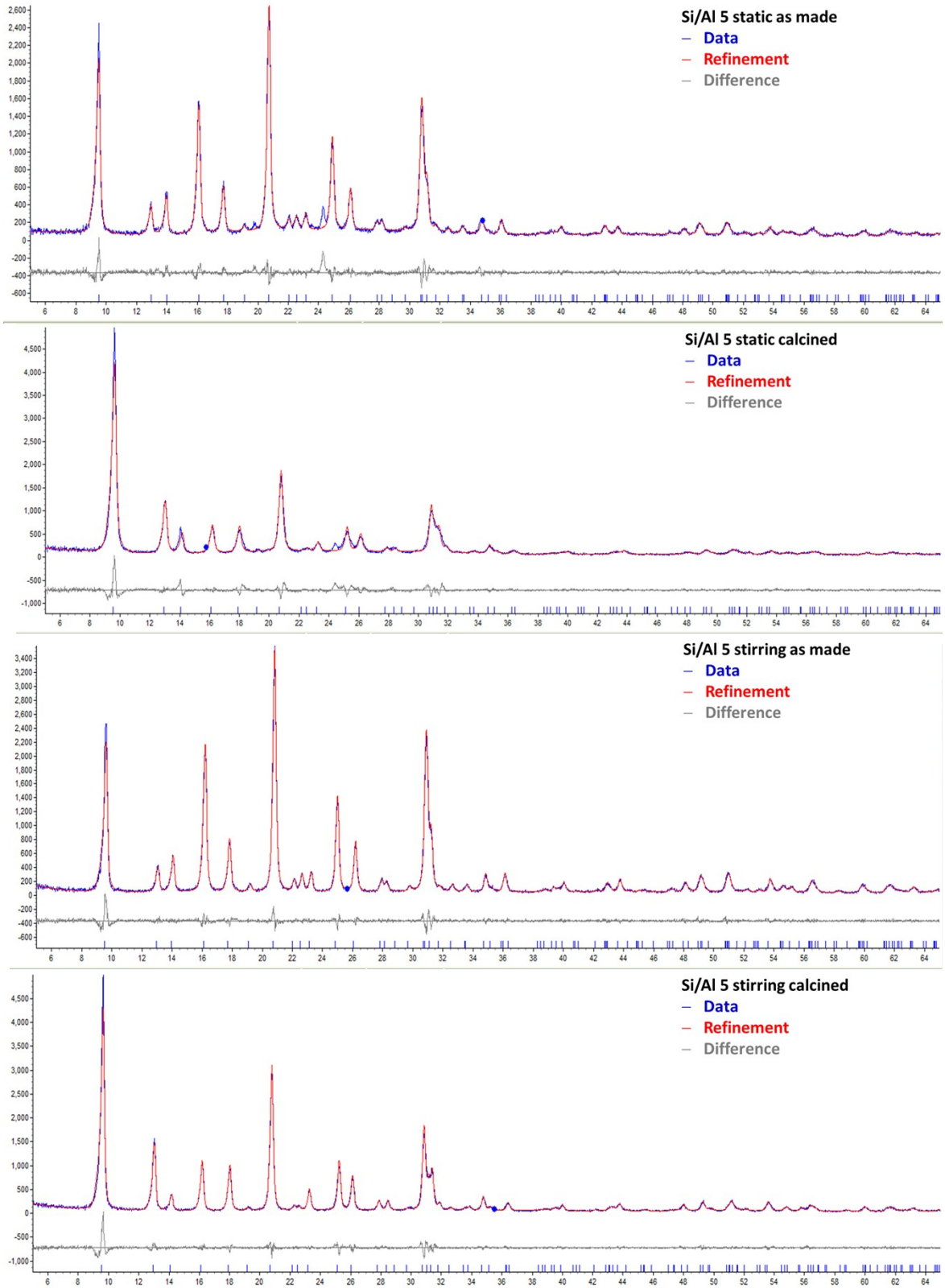


Figure S10. Refinement plots with experimental data (blue line), fit from refinement (red), and difference curve (grey) for the sample SSZ-13 (Si/Al 5) as made and calcined.

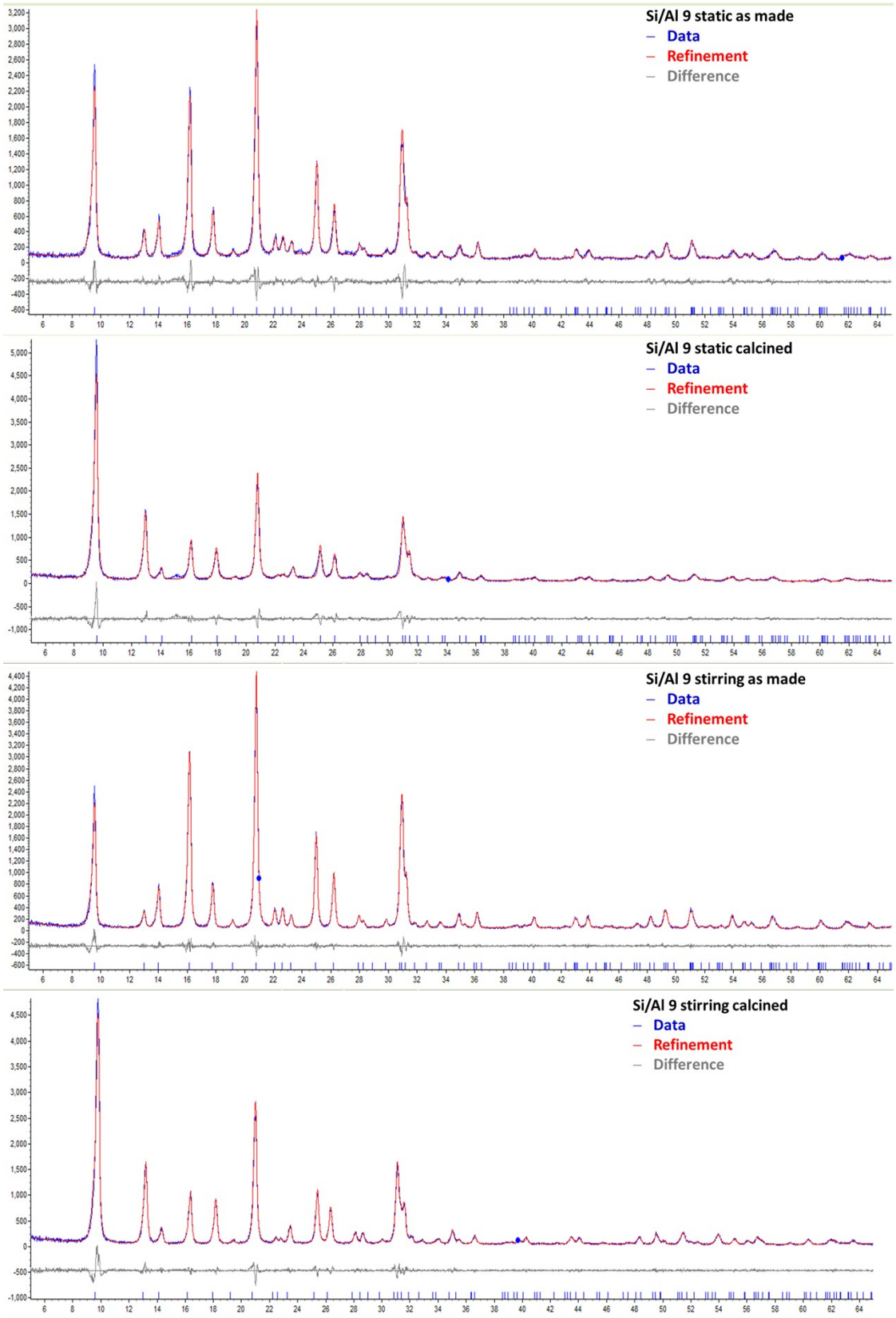


Figure S11. Refinement plots with experimental data (blue line), fit from refinement (red), and difference curve (grey) for the sample SSZ-13 (Si/Al 9) as made and calcined.

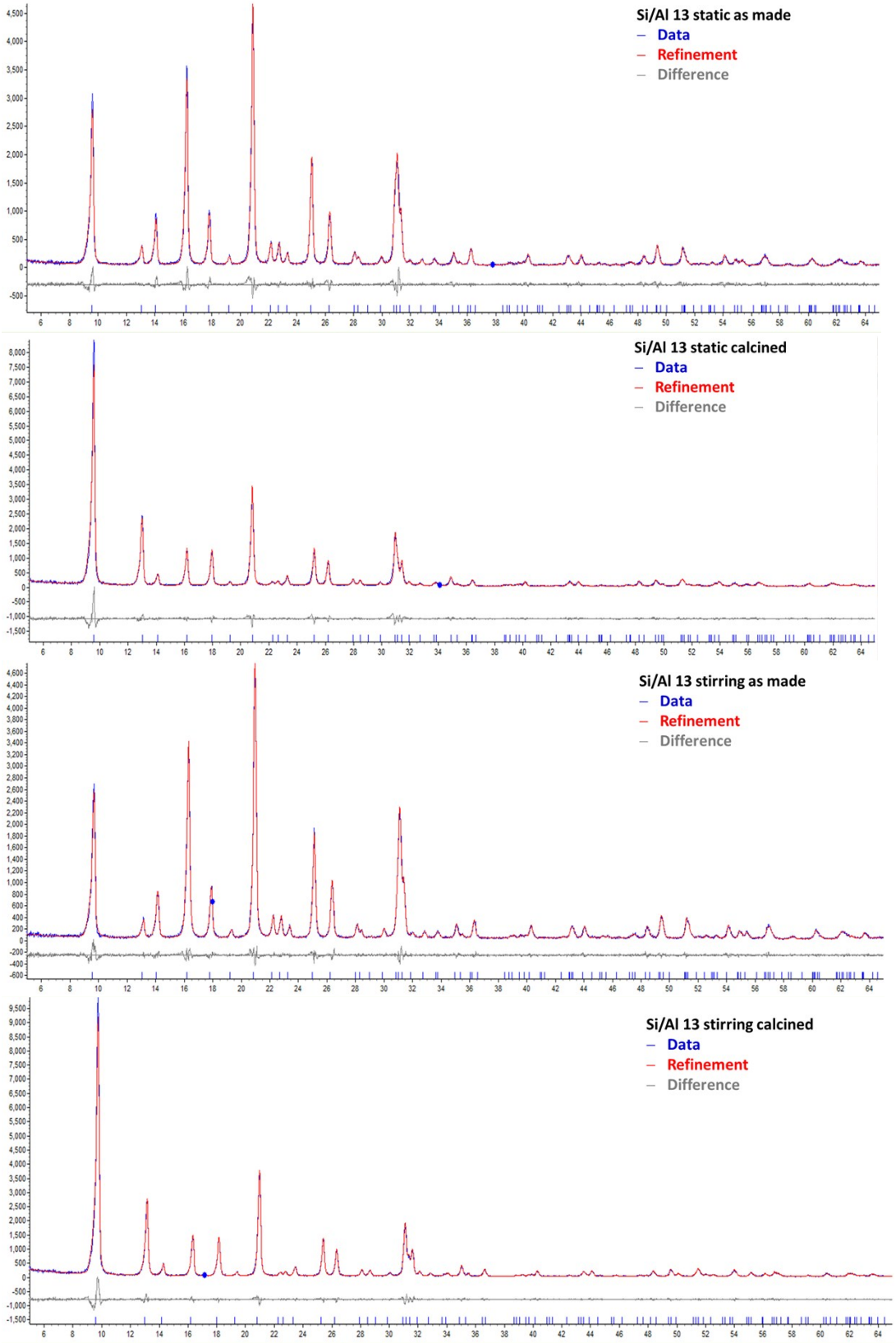


Figure S12. Refinement plots with experimental data (blue line), fit from refinement (red), and difference curve (grey) for the sample SSZ-13 (Si/Al 13) as made and calcined.

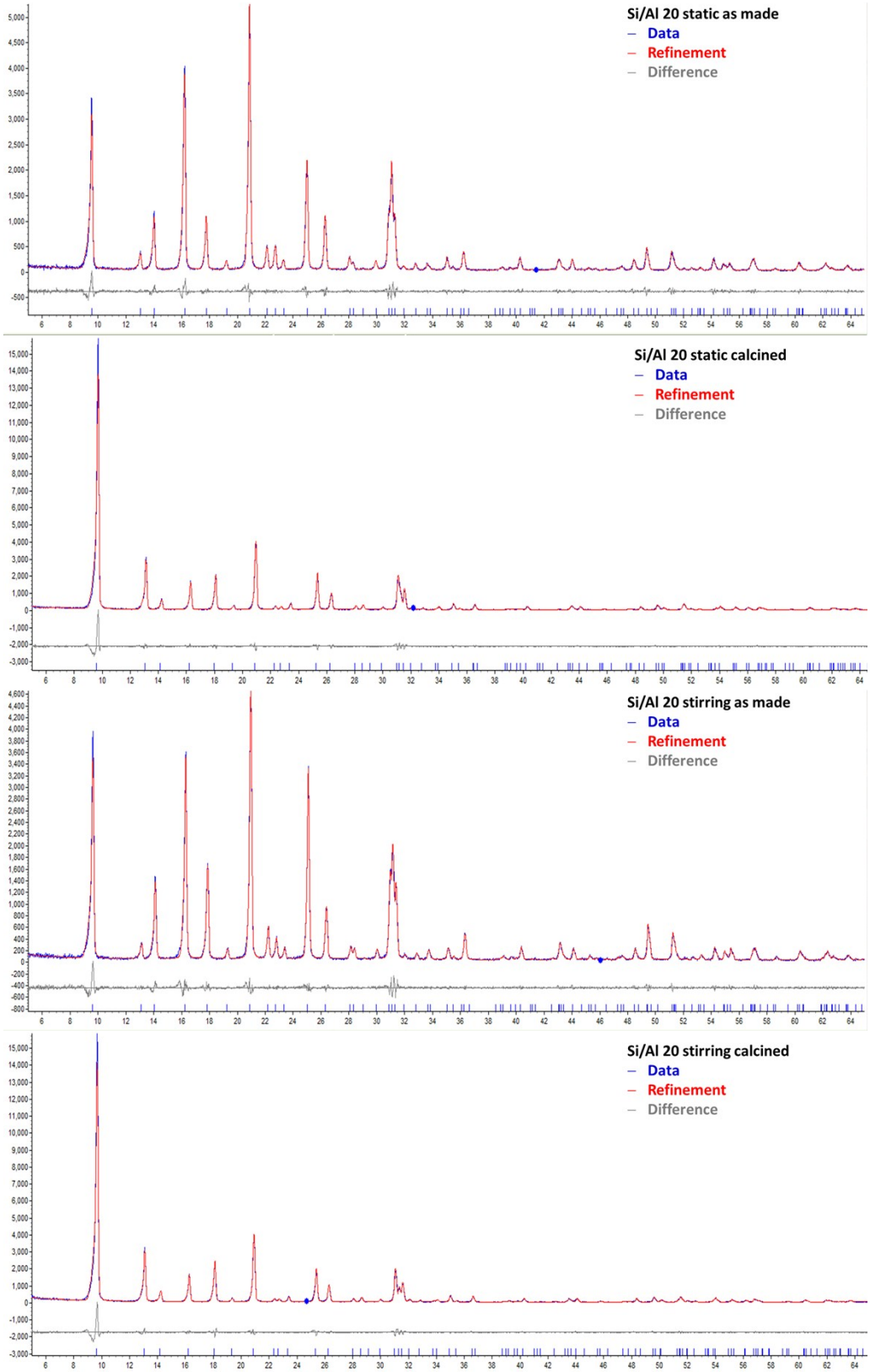


Figure S13. Refinement plots with experimental data (blue line), fit from refinement (red), and difference curve (grey) for the sample SSZ-13 (Si/Al 20) as made and calcined.