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**Supporting information for article:**

High-pressure intrusion of double salt aqueous solution in pure silica chabazite: searching for cation selectivity

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Thermogravimetric (TG) analyses were carried out on a Mettler Toledo STARe apparatus, under air flow, with a heating rate of 5 °C/min from 30 to 800 °C. $^{29}$Si MAS and $^{1}$H–$^{29}$Si CPMAS NMR spectra were recorded at room temperature on a Bruker Advance II 300 MHz spectrometer, with a double-channel 7 mm Bruker MAS probe. The recording conditions are given in Table SN.

Table S1. Recording conditions of the $^{29}$Si MAS NMR spectra.

|  | $^{29}$Si MAS |
|---|---|
| Chemical Shift Standard | Tetramethylsilane (TMS) |
| Frequency (MHz) | 59.6 |
| Pulse width (µs) | 2.3 |
| Flip angle | $\pi/6$ |
| Contact time (ms) | / |
| Recycle time (s) | 80 |
| Spinning rate (kHz) | 4 |
| Scans number | 1000 |
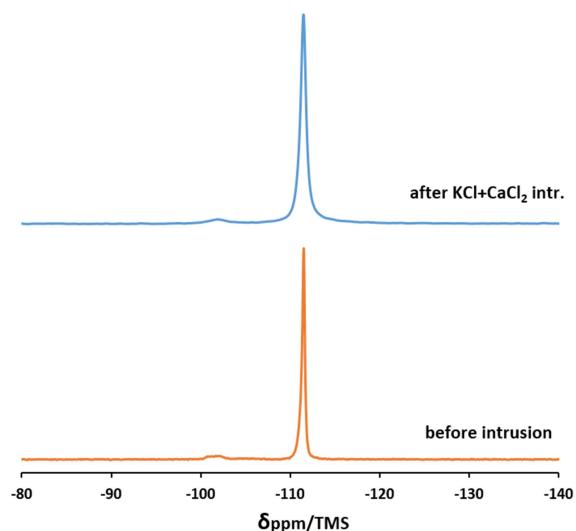


Figure S1. $^{29}$Si MAS NMR spectra of the Si-CHA samples before and after three intrusion–extrusion cycles in KCl 1M + CaCl$_2$ 1M aqueous solution.
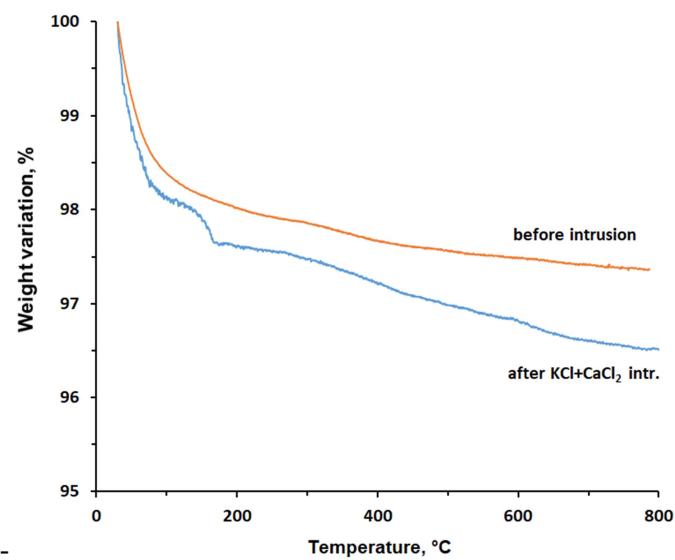
Figure S2. Thermogravimetric curves of the Si-CHA samples before and after three intrusion–extrusion cycles in KCl 1M + CaCl$_2$ 1M aqueous solution.

Table S2. atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA at ambient conditions.

|  | x | y | z | occ. | Biso (Å$^2$) |
|---|---|---|---|---|---|
| Si1 | 0.2291(1) | -0.0005(1) | 0.1042(1) | 1 | 1.00(5) |
| O1 | 0.1203(2) | -0.1203(2) | 0.1287(2) | 1 | 1.00(7) |
| O2 | 1/3 | 0.0190(3) | 1/6 | 1 | 1.00(7) |
| O3 | 0.2004(3) | 0.1002(2) | 0.1218(3) | 1 | 1.00(7) |
| O4 | 0.2622(3) | 0 | 0 | 1 | 1.00(7) |
| Wa | 0.514(5) | 0.257(3) | -0.262(6) | 0.08(1) | 15(3) |
| Wb | 0.215(5) | -0.215(5) | -0.638(9) | 0.09(1) | 15(3) |

Table S3. Evolution upon compression of the unit cell parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution.

| Pressure (GPa) | a (Å) | c (Å) | V (Å$^3$) |
|---|---|---|---|
| Ambient | 13.53759(1) | 14.76042(5) | 2342.68(1) |
| 0.14 | 13.5412(5) | 14.7482(5) | 2341.9(2) |
| 0.22 | 13.5364(9) | 14.7807(9) | 2345.5(4) |
| 0.38 | 13.5187(1) | 14.8081(1) | 2343.7(5) |
| 0.53 | 13.5013(9) | 14.8314(9) | 2341.3(9) |
| 0.74 | 13.4890(1) | 14.8388(9) | 2338.3(5) |
| 0.93 | 13.4871(1) | 14.8225(1) | 2335.0(5) |
| 1.17 | 13.5043(2) | 14.7301(9) | 2326.4(6) |

Table S4. Cell parameters, atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.14 GPa.

|     | x | y | z | occ. | Biso (Å$^2$) |
|-----|-----|-----|-----|-----|-----|
| Si1 | 0.2302(3) | -0.0014(4) | 0.1030(2) | 1 | 1.0(1) |
| O1  | 0.1189(4) | -0.1189(4) | 0.1262(6) | 1 | 1.0(2) |
| O2  | 1/3 | 0.0126(8) | 1/6 | 1 | 1.0(2) |
| O3  | 0.2069(8) | 0.1034(4) | 0.1224(7) | 1 | 1.0(2) |
| O4  | 0.2612(4) | 0 | 0 | 1 | 1.0(2) |
| Cl  | 0.911(2) | -0.911(2) | 0.336(3) | 0.050(2) | 7(2) |
| K   | 2/3 | 1/3 | -0.018(8) | 0.190(4) | 19(2) |
| Wa  | 0.486(3) | 0.243(1) | -0.218(2) | 0.255(6) | 20(1) |
| Wb  | 0.180(6) | -0.180(6) | -0.637(8) | 0.169(5) | 20(1) |

Table S5. Cell parameters, atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.22 GPa.

|     | x | y | z | occ. | Biso (Å$^2$) |
|-----|-----|-----|-----|-----|-----|
| Si1 | 0.2291(5) | 0.0004(6) | 0.1027(3) | 1 | 1.0(1) |
| O1  | 0.1176(6) | -0.1176(6) | 0.1268(7) | 1 | 1.0(2) |
| O2  | 1/3 | 0.017(1) | 1/6 | 1 | 1.0(2) |
| O3  | 0.2045(8) | 0.1023(4) | 0.1191(7) | 1 | 1.0(2) |
| O4  | 0.2621(3) | 0 | 0 | 1 | 1.0(2) |
| Cl  | 0.911(1) | -0.911(1) | 0.332(3) | 0.240(4) | 20.(1) |
| K   | 2/3 | 1/3 | 0.038(4) | 0.193(5) | 5.0(5) |
| Ca  | 0.132(8) | -0.132(8) | -0.71(2) | 0.087 | 19(2) |
| Wa  | 0.503(2) | 0.2514(8) | -0.226(1) | 0.666(6) | 20(1) |
| Wb  | 0.181(1) | -0.181(1) | -0.642(3) | 0.306(7) | 20(1) |

Table S6. Cell parameters, atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.38 GPa.

|     | x | y | z | occ. | Biso (Å$^2$) |
|-----|-----|-----|-----|-----|-----|
| Si1 | 0.2294(4) | -0.0006(5) | 0.1029(3) | 1 | 1.0(2) |
| O1  | 0.1184(5) | -0.1184(5) | 0.1261(7) | 1 | 1.0(3) |
| O2  | 1/3 | 0.0159(10) | 1/6 | 1 | 1.0(3) |
| O3  | 0.2042(8) | 0.1021(4) | 0.1199(8) | 1 | 1.0(3) |
| O4  | 0.2634(1) | 0 | 0 | 1 | 1.0(3) |
| Cl  | 0.9108(8) | -0.9108(8) | 0.332(2) | 0.295(4) | 20(1) |
| K   | 2/3 | 1/3 | 0.048(2) | 0.255(5) | 5(1) |

| | | | | | |
|---|---|---|---|---|---|
| Ca | 0.135(5) | -0.135(5) | -0.645(8) | 0.099 | 18(2) |
| Wa | 0.501(1) | 0.2545(6) | -0.228(1) | 0.666(6) | 11(1) |
| Wb | 0.179(9) | -0.179(9) | -0.642(8) | 0.342(7) | 11(1) |

Table S7. Cell parameters, atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.74 GPa.

| | x | y | z | occ. | Biso (Å$^2$) |
|---|---|---|---|---|---|
| Si1 | 0.2309(4) | -0.0011(4) | 0.1028(3) | 1 | 1.0(3) |
| O1 | 0.1187(5) | -0.1187(5) | 0.1252(7) | 1 | 1.0(3) |
| O2 | 1/3 | 0.013(1) | 1/6 | 1 | 1.0(3) |
| O3 | 0.2067(8) | 0.1033(4) | 0.1201(8) | 1 | 1.0(3) |
| O4 | 0.2659(1) | 0 | 0 | 1 | 1.0(3) |
| Cl | 0.9106(8) | -0.9106(8) | 0.332(2) | 0.310(5) | 19.7(9) |
| K | 2/3 | 1/3 | 0.055(2) | 0.333(6) | 4.9(9) |
| Ca | 0.135(6) | -0.135(6) | -0.641(9) | 0.099 | 13(2) |
| Wa | 0.5095(9) | 0.2547(5) | -0.2285(9) | 0.666(7) | 6.5(8) |
| Wb | 0.179(10) | -0.179(10) | -0.643(8) | 0.406(8) | 6.5(8) |

Table S8. Cell parameters, atomic coordinates, occupancy factors and isotropic thermal parameters of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.93 GPa.

| | x | y | z | occ. | B (Å$^2$) |
|---|---|---|---|---|---|
| Si1 | 0.2311(5) | -0.0012(4) | 0.1029(3) | 1 | 1.0(2) |
| O1 | 0.1181(5) | -0.1181(5) | 0.1250(7) | 1 | 1.0(3) |
| O2 | 1/3 | 0.01158 | 1/6 | 1 | 1.0(3) |
| O3 | 0.2086(8) | 0.1043(4) | 0.1207(8) | 1 | 1.0(3) |
| O4 | 0.2658(9) | 0 | 0 | 1 | 1.0(3) |
| Cl | 0.9106(8) | -0.9106(8) | 0.3324(18) | 0.310(4) | 12.8(8) |
| K | 2/3 | 1/3 | 0.0563(16) | 0.333(6) | 1.3(8) |
| Ca | 0.135(10) | -0.135(10) | -0.646(19) | 0.099 | 12(2) |
| Wa | 0.5095(9) | 0.2548(6) | -0.2286(9) | 0.666(8) | 6.3(8) |
| Wb | 0.179(9) | -0.179(9) | -0.643(8) | 0.402(8) | 6.3(8) |
| Wc | 0 | 0 | 0 | 0.050(2) | 6.3(8) |

Table S9. Framework and extraframework bond distances (<3.5 Å) for the refined structures

| | | | P amb | 0.14 GPa | 0.22 GPa | 0.38 GPa | 0.74 GPa | 0.93 GPa |
|---|---|---|---|---|---|---|---|---|
| **Si1** | **O1** | | 1.594(2) | 1.591(3) | 1.595(6) | 1.586(5) | 1.586(5) | 1.585(5) |
| | **O2** | | 1.593(2) | 1.607(4) | 1.613(7) | 1.613(7) | 1.603(7) | 1.608(5) |
| | **O3** | | 1.615(2) | 1.617(5) | 1.597(9) | 1.606(8) | 1.617(8) | 1.619(8) |
| | **O4** | | 1.601(2) | 1.584(2) | 1.584(6) | 1.590(6) | 1.594(4) | 1.592(6) |
| | | | | | | | | |
| **Wa** | **O2** | x2 | | 3.17(2) | 3.36(2) | 3.20(2) | 3.36(1) | 3.33(1) |
| | **Cl** | | | 3.17(3) | 3.38(4) | 2.60(8) | 3.43(3) | 3.41(3) |
| | **K** | x3 | | 2.72(5) | 2.86(2) | 2.91(1) | 3.01(3) | 3.01(2) |
| | **Wa** | x2 | 3.10(1) | 2.60(5) | 2.60(3) | 2.60(2) | 2.60(2) | 2.60(2) |
| | **Wb** | x2 | 3.31(1) | 2.60(5) | 2.6(3) | 2.60(8) | 2.60(2) | 2.60(9) |
| | | | | | | | | |
| **Wb** | **O1** | | 2.60(9) | 2.71(10) | 2.8(4) | 2.83(13) | 2.86(9) | 2.8(1) |
| | **O2** | x2 | | 3.11(6) | 3.2(3) | 3.17(8) | 3.17(9) | 3.15(8) |
| | **O4** | x2 | | 3.27(3) | 3.25(6) | 3.23(3) | 3.19(3) | 3.19(4) |
| | **Cl** | x2 | | 3.15(9) | 3.2(2) | 3.2(2) | 3.15(19) | 3.15(9) |
| | **Wa** | | 3.31(2) | 2.60(5) | 2.6(3) | 2.60(8) | 2.60(2) | 2.60(9) |
| | | | | | | | | |
| **Cl** | **O1** | | | | 3.51(3) | | | |
| | **O2** | | | | | | 3.67(3) | |
| | **O3** | x2 | | 3.17(1) | 3.16(4) | 3.16(4) | 3.16(3) | 3.16(3) |
| | **O4** | x2 | | 3.20(5) | 3.19(2) | 3.17(8) | 3.15(2) | 3.15(2) |
| | **Wb** | x2 | | 3.15(9) | 3.2(2) | 3.2(2) | 3.15(19) | 3.15(9) |
| | **Wa** | | | 3.17(3) | 3.38(4) | 3.41(4) | 3.43(3) | 3.41(3) |
| | **Ca** | | | | 2.8(1) | 2.80(9) | 2.80(9) | 2.80(9) |
| | | | | | | | | |
| **K** | **Wa** | x3 | | 2.72(5) | 2.86(2) | 2.91(1) | 3.01(3) | 3.01(2) |
| | | | | | | | | |
| **Ca** | **O1** | | | | 2.4(4) | | | |
| | **Cl** | | | | 2.8(2) | 2.80(9) | 2.80(9) | 2.80(9) |
| | | | | | | | | |
| **Wc** | **O3** | | | | | | | 3.02(1) |

Table S10. Details of structural refinement parameters of Si-CHA under intrusion of Si-CHA at ambient conditions.

| | **Pamb** | **0.14 GPa** | **0.22 GPa** | **0.38 GPa** | **0.74 GPa** | **0.93 GPa** |
|---|---|---|---|---|---|---|
| **Rwp (%)** | 10.6 | 2.13 | 2.52 | 2.23 | 2.25 | 2.32 |
| **$R_{exp}$ (%)** | 3.56 | 2.56 | 2.68 | 2.68 | 2.67 | 2.68 |
| **Rp (%)** | 8 | 1.06 | 1.36 | 1.19 | 1.17 | 1.21 |
| **No. of variables** | 50 | 50 | 50 | 50 | 50 | 50 |
| **No. of reflections** | 422 | 233 | 233 | 233 | 233 | 233 |
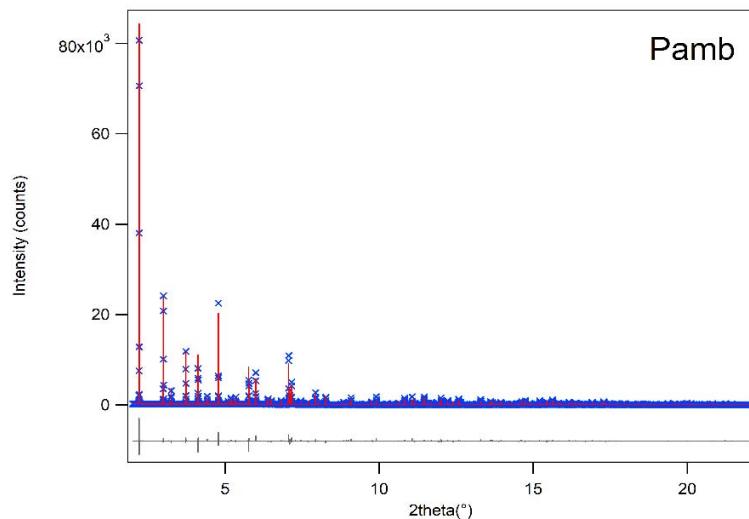


Figure S3. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA at ambient conditions.
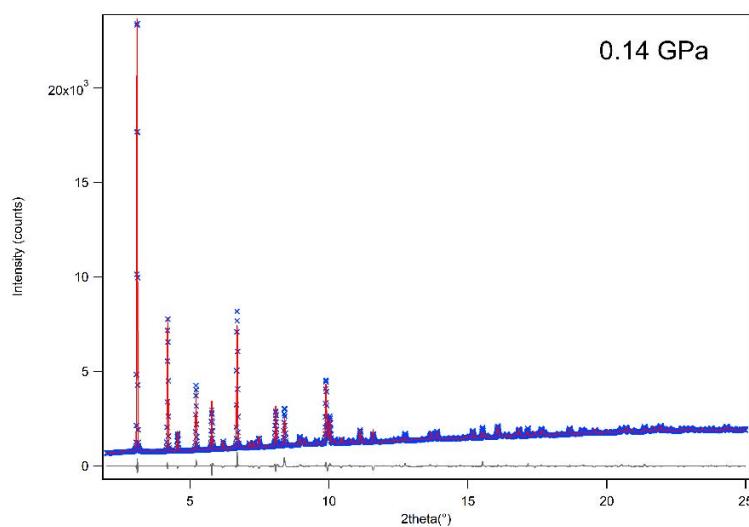


Figure S4. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA under intrusion of KCl 1M + $CaCl_2$ 1M aqueous solution at 0.22 GPa.
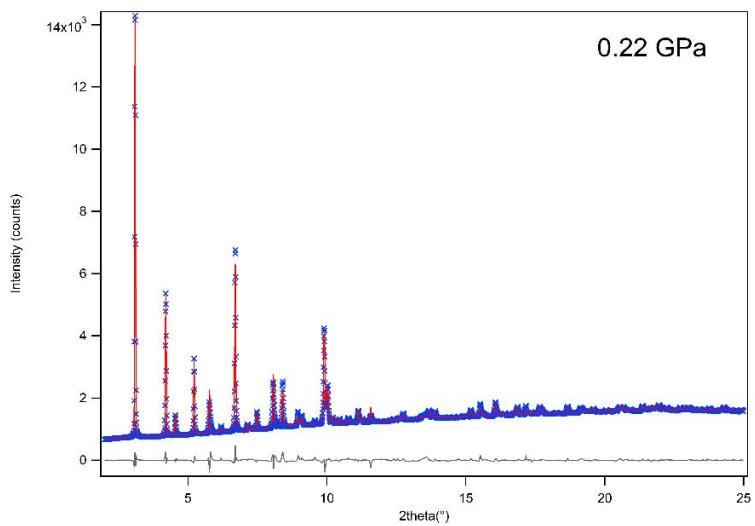
Figure S5. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.22 GPa.
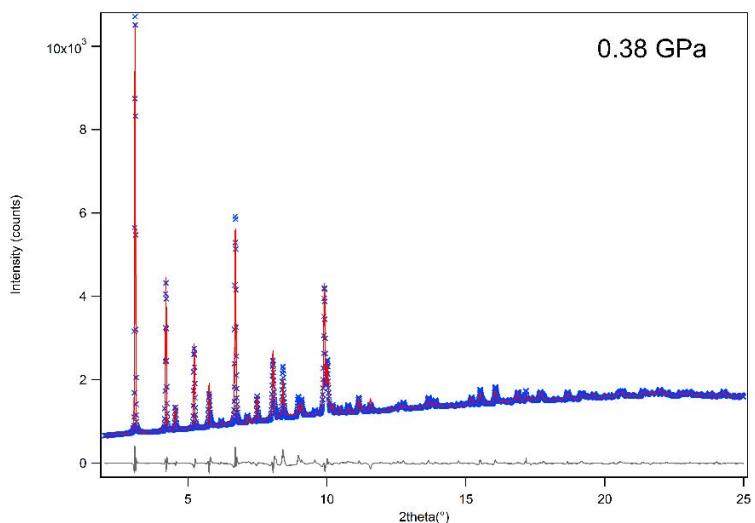


Figure S6. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.38 GPa.
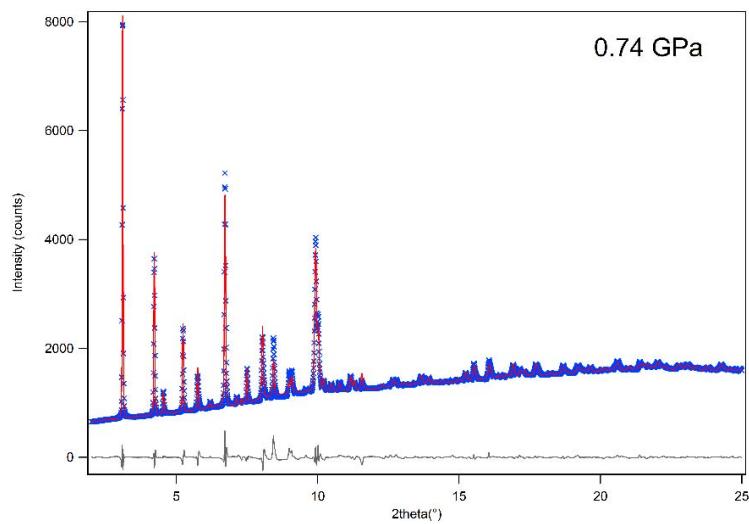
Figure S7. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.74 GPa.
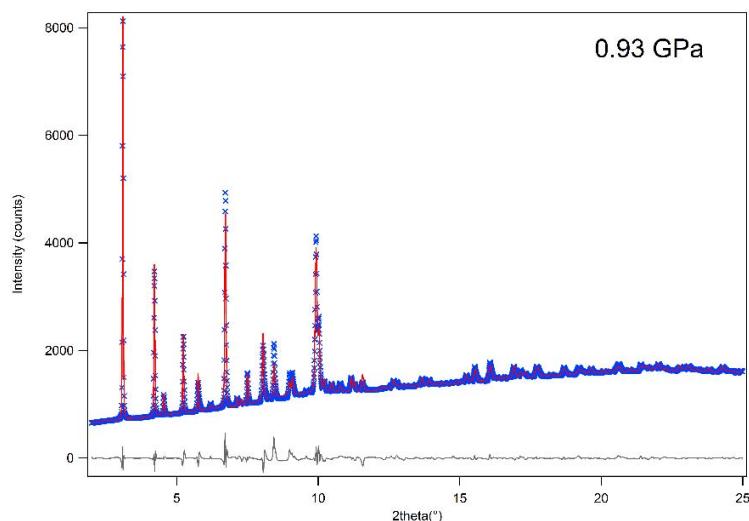


Figure S8. Observed (blue marks) and calculated (red line) diffraction patterns and final difference curve (grey line) from Rietveld refinements of Si-CHA under intrusion of KCl 1M + CaCl$_2$ 1M aqueous solution at 0.93 GPa.