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Supporting information for article:

New zeolite-like RUB-5 and its related hydrous layer silicate RUB-6 structurally characterised by electron microscopy

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S1. Crystal images

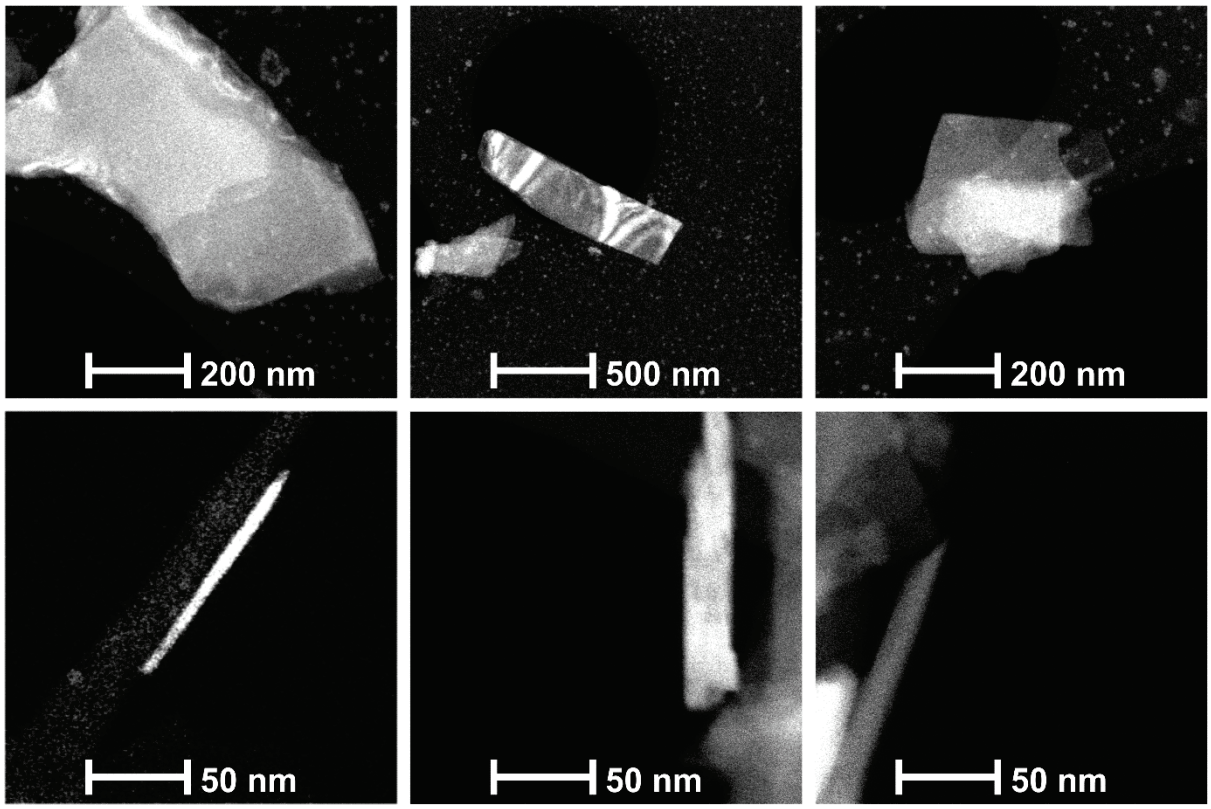

Figure S1 µm-probe STEM images of RUB-5 collected with low electron dose (C2 of 10µm, SS8 and GL8) showing the plate-like morphology (upper part). The plate thickness of the crystals is shown (lower part) by upstanding crystals at the holes of the carbon film.

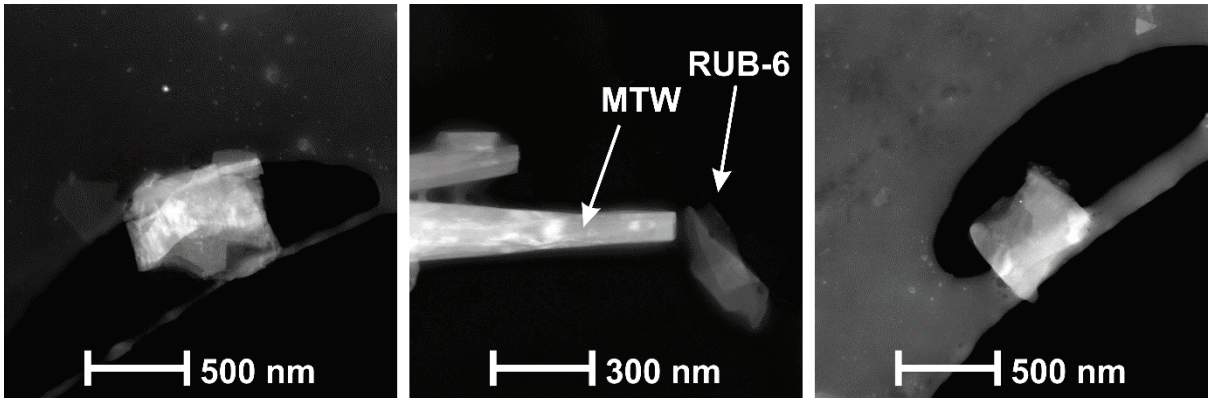

Figure S2 µm-probe STEM images of RUB-6 showing highly agglomerated plate-like crystals. Images are recorded with much higher electron dose (C2 of 50µm, SS8 and GL8) than for RUB-5 because of low scattering power of RUB-6. Zeolite MTW in comparison with RUB-6 (middle).

S2. Energy-dispersive X-ray spectroscopy (EDXS)

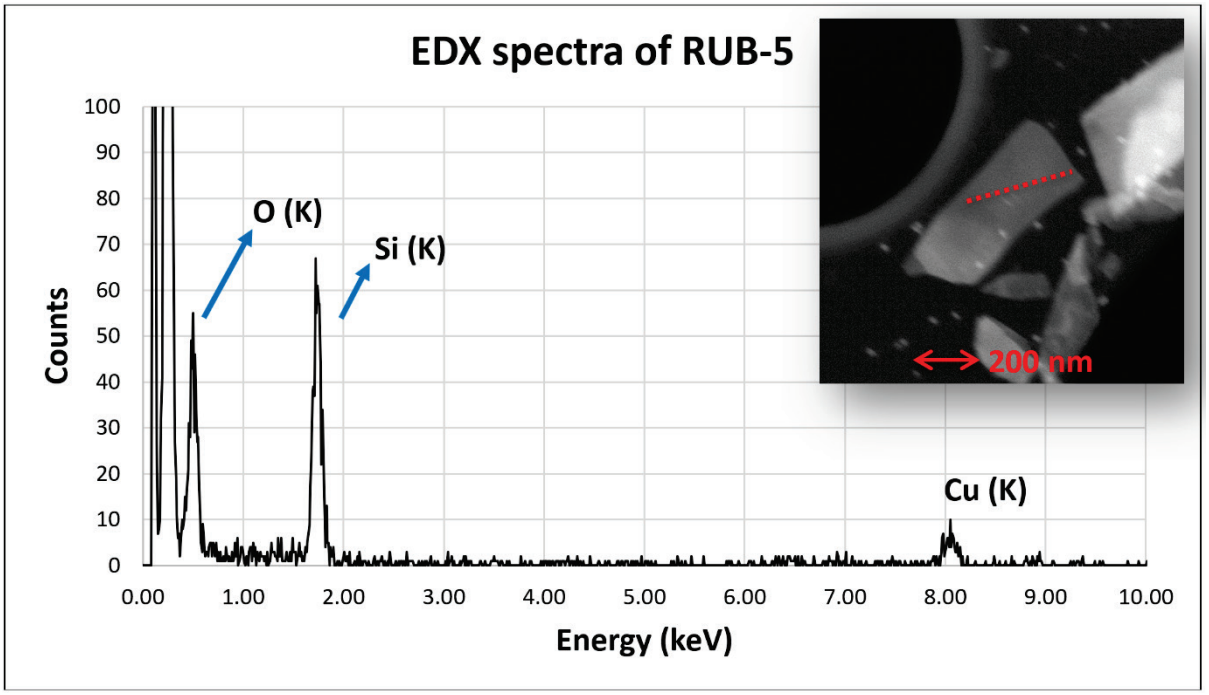

Figure S3 EDX spectrum of RUB-5 particle shown in STEM image with the most intense fluorescence peaks labelled. The Cu K peak at about 8 keV is caused by the TEM Grid.

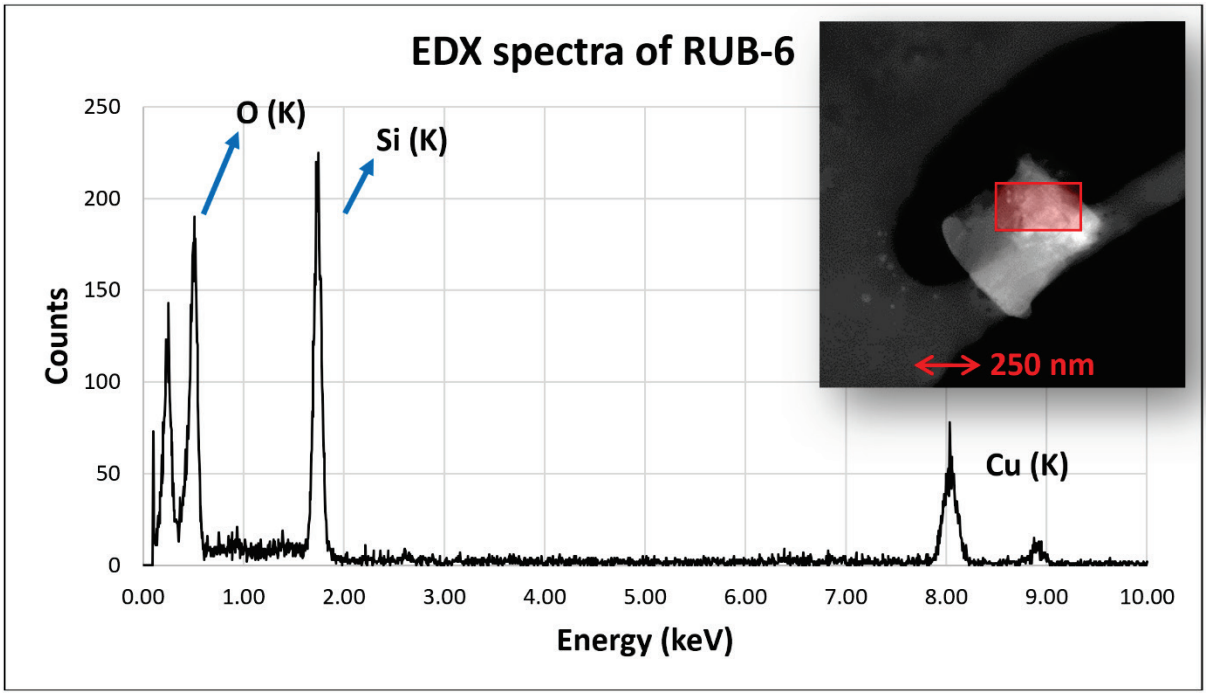

Figure S4 EDX spectrum of RUB-6 particle shown in STEM image with the most intense fluorescence peaks labelled. The Cu K peak at about 8 keV is caused by the TEM Grid.

S3. cif data determined by ADT

S3.1. RUB-5

```
#######################################################################
#
# Structure solution of RUB-5
#
#######################################################################

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_symmetry_equiv_pos_as_xyz
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2 -x,y,-z
3 1/2+x,1/2+y,z
4 1/2-x,1/2+y,-z
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_cell_length_b                   10.64
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_cell_angle_beta                 106.4
_cell_angle_gamma                90
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 _atom_site_fract_z
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 _atom_site_U_iso_or_equiv
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  Si10 Si 0.86695 0.91446 0.24052 Uiso 0.0214 4 1 d . . .
  Si11 Si 0.70398 0.71956 0.12492 Uiso 0.0445 4 1 d . . .
  Si12 Si 0.17398 0.63515 0.24603 Uiso 0.0227 4 1 d . . .
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  O2 O 0.2653 0.98759 0.27036 Uiso 0.0229 4 1 d . . .
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  O9 O 0.98182 0.16435 0.42375 Uiso 0.026 4 1 d . . .
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  O11 O 0.42422 0.94737 0.05741 Uiso 0.0229 4 1 d . . .
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  O15 O 0.29076 0.51476 0.26016 Uiso 0.0333 4 1 d . . .
  O16 O 0.43172 0.8991 0.19032 Uiso 0.0261 4 1 d . . .
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#END
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S3.2. RUB-6

```
######################################################################
#
# Structure solution of RUB-6
#
######################################################################

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_symmetry_Int_Tables_number      5
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_symmetry_equiv_pos_as_xyz
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2 -x,y,-z
3 1/2+x,1/2+y,z
4 1/2-x,1/2+y,-z
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_cell_length_b                   10.67
_cell_length_c                   20.56
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  Si7 Si 0.84568 0.04209 0.71095 Uiso 0.0521 4 1 d . . .
  Si8 Si 0.77085 0.20016 0.8277 Uiso 0.0741 4 1 d . . .
  O1 Si 0.62541 0.8532 0.70562 Uiso 0.0712 4 1 d . . .
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  O4 O 0.45207 0.29661 0.3458 Uiso 0.0986 4 1 d . . .
  Si11 O 0.49643 0.12072 0.42744 Uiso 0.0712 4 1 d . . .
  O5 O 0.00699 0.30576 0.64432 Uiso 0.0704 4 1 d . . .
  O6 Si 0.79916 0.62699 0.72218 Uiso 0.022 4 1 d . . .
  O7 O 0.85945 0.13516 0.65709 Uiso 0.1222 4 1 d . . .
  O8 O 0.31219 0.4956 0.29747 Uiso 0.0423 4 1 d . . .
```

```
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O14 O 0.83906 0.1436 0.7743 Uiso 0.0751 4 1 d . . .
O15 O 0.87485 0.47005 0.50163 Uiso 0.083 4 1 d . . .
O16 O 0.72079 0.72199 0.72736 Uiso 0.0975 4 1 d . . .
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O18 O 0.90249 0.27822 0.90013 Uiso 0.1387 4 1 d . . .
O19 O 0.29128 0.34137 0.22564 Uiso 0.1015 4 1 d . . .
O21 O 0.50157 0.94996 0.89416 Uiso 0.0141 4 1 d . . .
O23 O 0.40622 0.06345 0.78849 Uiso 0.0742 4 1 d . . .
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Si6 Si 0 0.07144 0.5 Uiso 0.0524 2 1 d . . .

#END
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S4. Rietveld refinements

S4.1. Rietveld plots

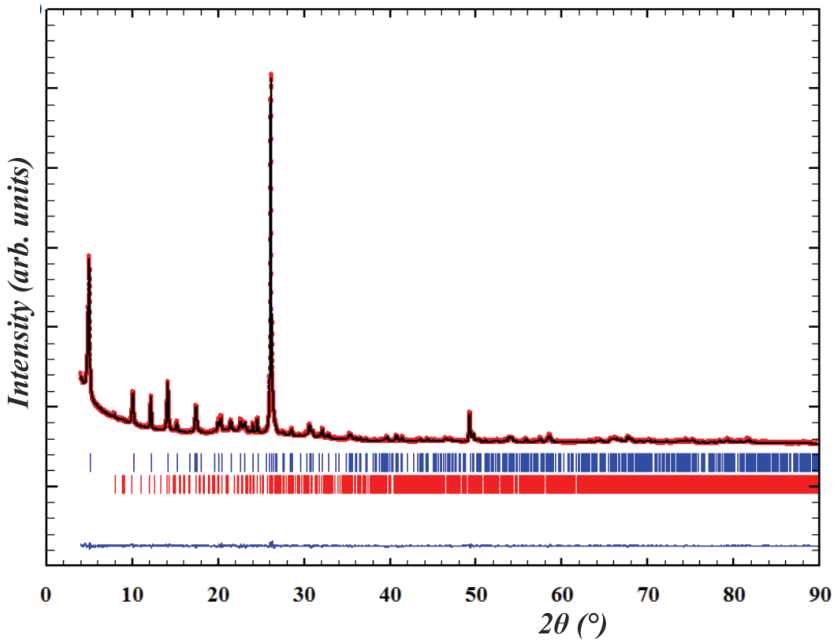

Figure S5 Plot of the diffraction patterns after Rietveld analysis of RUB-5. Experimental (red) and calculated (black) patterns are shown above the difference plot (blue line). Tick marks for allowed reflections (blue = RUB-5, red = ZSM-5) are given. Intensities in arbitrary units.

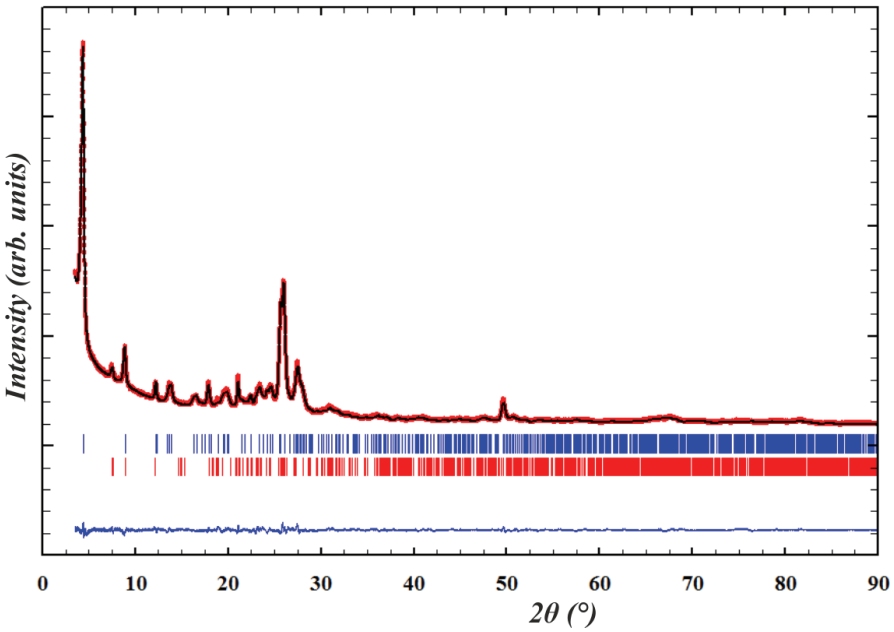

Figure S6 Plot of the diffraction patterns after Rietveld analysis of RUB-6. Experimental (red) and calculated (black) patterns are shown above the difference plot (blue line). Tick marks for allowed reflections (blue = RUB-6, red = ZSM-12) are given. Intensities in arbitrary units.

S4.2. Parameters for the Rietveld refinements

Table S1 Experimental and crystallographic parameters for the Rietveld refinements of zeolite RUB-5 and layer silicate RUB-6.

	RUB-5	RUB-6
Diffractometer	Siemens D5000 with 6° Braun PSD	
Wavelength	1.54060 Å	
Sample	powder in 0.3 mm glass capillary	
2θ range (°)	4.0 - 90.0	3.5 - 90.0
Step size in 2θ (°)	0.007866	0.007888
No. steps	11387	11007
No. contributing reflections	833	928
No. geometric restraints	129	135
No. structural parameters	102	113
No. profile parameters	21	21
FWHM at ca. 25° 2θ (°)	0.12 - 0.32	0.22 - 0.82
Spherical harmonics component Y00	0.194598	5.397610
Spherical harmonics component Y20	-0.977645	-0.950710

Spherical harmonics component Y22+	1.146215	2.325654
Spherical harmonics component Y22-	-0.702390	-1.144377
Spherical harmonics component Y40	-0.079124	-0.555880
Spherical harmonics component Y42+	1.086668	5.505152
Spherical harmonics component Y42-	-1.325602	1.866171
Spherical harmonics component Y44+	-0.742849	-1.067283
Impurity in weight percent	1.6(2)% of ZSM-5	6.7(3)% of ZSM-12
R_{Bragg}	0.033	0.026
R_F	0.031	0.025
R_{wp}	0.086	0.089
R_{exp}	0.048	0.047
$\chi 2$	3.3	3.7
Space group	*C2* (No. 5)	*C2* (No. 5)
a_0 (Å)	10.2703 (6)	10.1250 (24)
b_0 (Å)	10.6423 (6)	10.6669 (26)
c_0 (Å)	18.1565 (6)	20.5551 (38)
β (°)	106.35 (2)	105.86 (1)
V_{UC} (Å^3)	1904.3 (2)	2135 (2)
Calculated density (g/cm^3)	2.20	2.01
Unit cell content	$[Si_{42}O_{84}]$	$[Si_{40}O_{76}(OH)_8]$ * $2C_6H_{14}N_2$

S4.3. Schematic drawing of the structure RUB-6

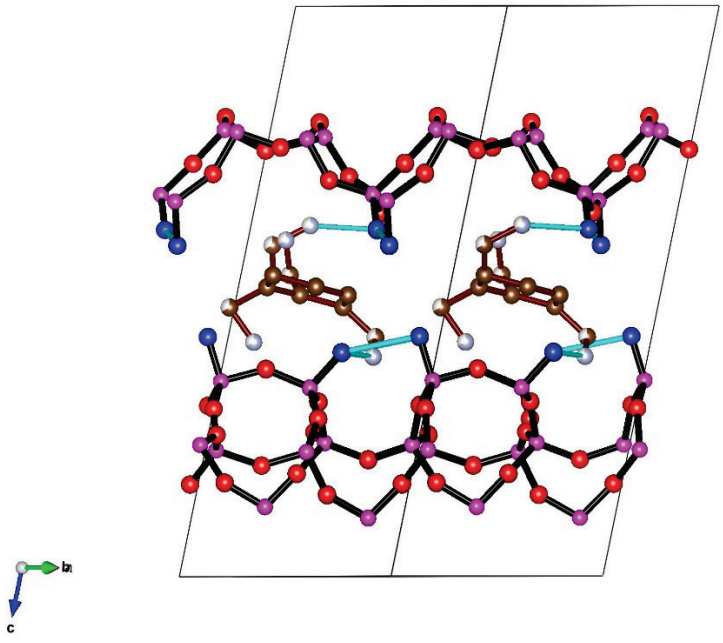

Figure S7 Section of the structure of RUB-6 showing the position of the organic molecule/cation in the interlayer region (projection along [-110]) (purple = Si, red = O, blue = terminal OH groups, brown = C, gray = N). Blue lines = hydrogen bonds.

S4.4. Atomic coordinates

Table S2 Atomic coordinates, displacement parameters and occupancy factors of the average structure of RUB-5.

Atom	x	y	z	U_{iso} (Å²)	Occ.
SI1	0.50000	0.1615(22)	0.50000	0.012	1.00000
SI2	0.4285(15)	0.0102(15)	0.6263(11)	0.012	1.00000
SI3	0.00000	0.9418(22)	0.00000	0.012	1.00000
SI4	0.2519(15)	0.6131(15)	0.0095(7)	0.012	1.00000
SI5	0.2839(15)	0.2815(15)	0.3717(7)	0.012	1.00000
SI6	0.00000	0.4412(22)	0.00000	0.012	1.00000
SI7	0.1496(15)	0.6396(15)	0.2507(7)	0.012	1.00000
SI8	0.0638(19)	0.7751(15)	0.1203(7)	0.012	1.00000
SI9	0.3708(15)	0.4327(15)	0.2502(11)	0.012	1.00000
SI10	0.0562(15)	0.2742(15)	0.1332(7)	0.012	1.00000
SI11	0.1185(15)	0.1017(15)	0.2532(11)	0.012	1.00000
SI12	0.6726(15)	0.1236(15)	0.7417(7)	0.012	1.00000
O1	0.2314(34)	0.5176(37)	0.2314(22)	0.024	1.00000
O2	0.1856(37)	0.1879(45)	0.0880(19)	0.024	1.00000
O3	0.3811(45)	0.3600(41)	0.1774(19)	0.024	1.00000
O4	0.0180(37)	0.8581(45)	0.0739(19)	0.024	1.00000
O5	0.4963(37)	0.5285(45)	0.2725(22)	0.024	1.00000
O6	0.3753(41)	0.2452(49)	0.4553(15)	0.024	1.00000
O7	0.0692(34)	0.1985(45)	0.1860(22)	0.024	1.00000
O8	0.0069(49)	0.3584(45)	0.0730(19)	0.024	1.00000
O9	0.1051(45)	0.3602(37)	0.1919(19)	0.024	1.00000
O10	0.1830(37)	0.7188(34)	0.0504(19)	0.024	1.00000

O11	0.2573(52)	0.7527(37)	0.2843(19)	0.024	1.00000
O12	0.1853(45)	0.1688(37)	0.3334(19)	0.024	1.00000
O13	0.0362(41)	0.6739(37)	0.1748(19)	0.024	1.00000
O14	0.3687(37)	0.3389(37)	0.3180(19)	0.024	1.00000
O15	0.1375(37)	0.5277(45)	0.0302(19)	0.024	1.00000
O16	0.1373(37)	1.0243(49)	0.0160(19)	0.024	1.00000
O17	0.2317(41)	0.0033(30)	0.2378(19)	0.024	1.00000
O18	0.4672(56)	0.0674(41)	0.5612(19)	0.024	1.00000
O19	0.1970(41)	0.3925(37)	0.3983(22)	0.024	1.00000
O20	0.0800(41)	0.5970(34)	0.3143(19)	0.024	1.00000
O21	0.5661(37)	0.0887(45)	0.6625(15)	0.024	1.00000
OH1	0.363(11)	0.484(11)	0.5296(56)	0.024	0.562(9)
OH2	0.50000	0.5840(64)	0.50000	0.024	1.00000
OH3	0.1820(64)	0.0358(64)	0.1282(30)	0.024	1.00000

Note: Realistic s.u. were obtained by multiplying the calculated s.u. with a factor of 3.74 (as provided by the FullProf program after the final stage of the Rietveld refinement).

Table S3 Atomic coordinates, displacement parameters and occupancy factors of the average structure of RUB-6.

Atom	x	y	z	U_{iso} (Å²)	Occ.
SI2	0.4437(23)	0.0091(19)	0.6690(9)	0.012	1.00000
SI3	0.00000	0.9459(28)	0.00000	0.012	1.00000
SI4	0.2614(23)	0.6257(19)	0.0153(9)	0.012	1.00000
SI5	0.3050(23)	0.3142(19)	0.3415(9)	0.012	1.00000
SI6	0.00000	0.4457(28)	0.00000	0.012	1.00000
SI7	0.1508(23)	0.6319(23)	0.2169(9)	0.012	1.00000
SI8	0.0631(23)	0.7720(19)	0.1026(9)	0.012	1.00000
SI9	0.3929(23)	0.4494(19)	0.2207(9)	0.012	1.00000
SI10	0.0428(23)	0.2946(19)	0.1209(9)	0.012	1.00000
SI11	0.1444(23)	0.1186(19)	0.2301(9)	0.012	1.00000

SI12	0.6887(23)	0.1266(19)	0.7671(9)	0.012	1.00000
O1	0.2482(51)	0.5285(61)	0.1947(28)	0.024	1.00000
O2	0.1632(61)	0.1948(61)	0.0840(23)	0.024	1.00000
O3	0.3895(66)	0.3529(61)	0.1601(28)	0.024	1.00000
O4	0.0218(47)	0.8611(61)	0.0657(23)	0.024	1.00000
O5	0.5132(47)	0.5518(56)	0.2475(28)	0.024	1.00000
O6	0.3791(80)	0.2893(70)	0.4200(24)	0.024	1.00000
O7	0.0769(56)	0.2023(66)	0.1638(28)	0.024	1.00000
O8	0.0338(47)	0.3618(66)	0.0692(23)	0.024	1.00000
O9	0.0991(66)	0.3569(66)	0.1794(28)	0.024	1.00000
O10	0.1913(51)	0.7184(51)	0.0447(28)	0.024	1.00000
O11	0.2504(75)	0.7460(42)	0.2528(23)	0.024	1.00000
O12	0.2215(75)	0.1952(51)	0.2991(23)	0.024	1.00000
O13	0.0462(61)	0.6674(51)	0.1435(19)	0.024	1.00000
O14	0.3844(56)	0.3858(66)	0.2925(23)	0.024	1.00000
O15	0.1300(51)	0.5406(61)	0.0133(37)	0.024	1.00000
O16	0.1305(51)	1.0326(61)	0.0040(37)	0.024	1.00000
O17	0.2241(75)	0.0030(47)	0.2057(23)	0.024	1.00000
O18	0.4772(80)	0.0767(51)	0.6096(28)	0.024	1.00000
O19	0.1954(51)	0.4072(66)	0.3613(23)	0.024	1.00000
O20	0.0838(70)	0.5875(51)	0.2764(28)	0.024	1.00000
O21	0.5816(51)	0.0922(61)	0.6950(19)	0.024	1.00000
C1	0.6775(47)	0.8348(33)	0.5264(33)	0.051	1.33333
C3	0.5805(33)	0.2838(47)	0.5059(42)	0.051	1.33333
C5	0.5795(28)	0.9498(42)	0.5064(37)	0.051	1.33333
C2	0.624(19)	0.601(5)	0.4718(33)	0.051	0.66667
N2	0.529(14)	0.656(14)	0.4117(18)	0.051	0.66667

Note: Realistic s.u. were obtained by multiplying the calculated s.u. with a factor of 4.68 (as provided by the FullProf program after the final stage of the Rietveld refinement). The occupancy factors of C and N atoms are increased to account for the hydrogen atoms bonded to C and N.

S4.5. cif-files

Please see attached files.

1. Structure of RUB-5 polymorph II

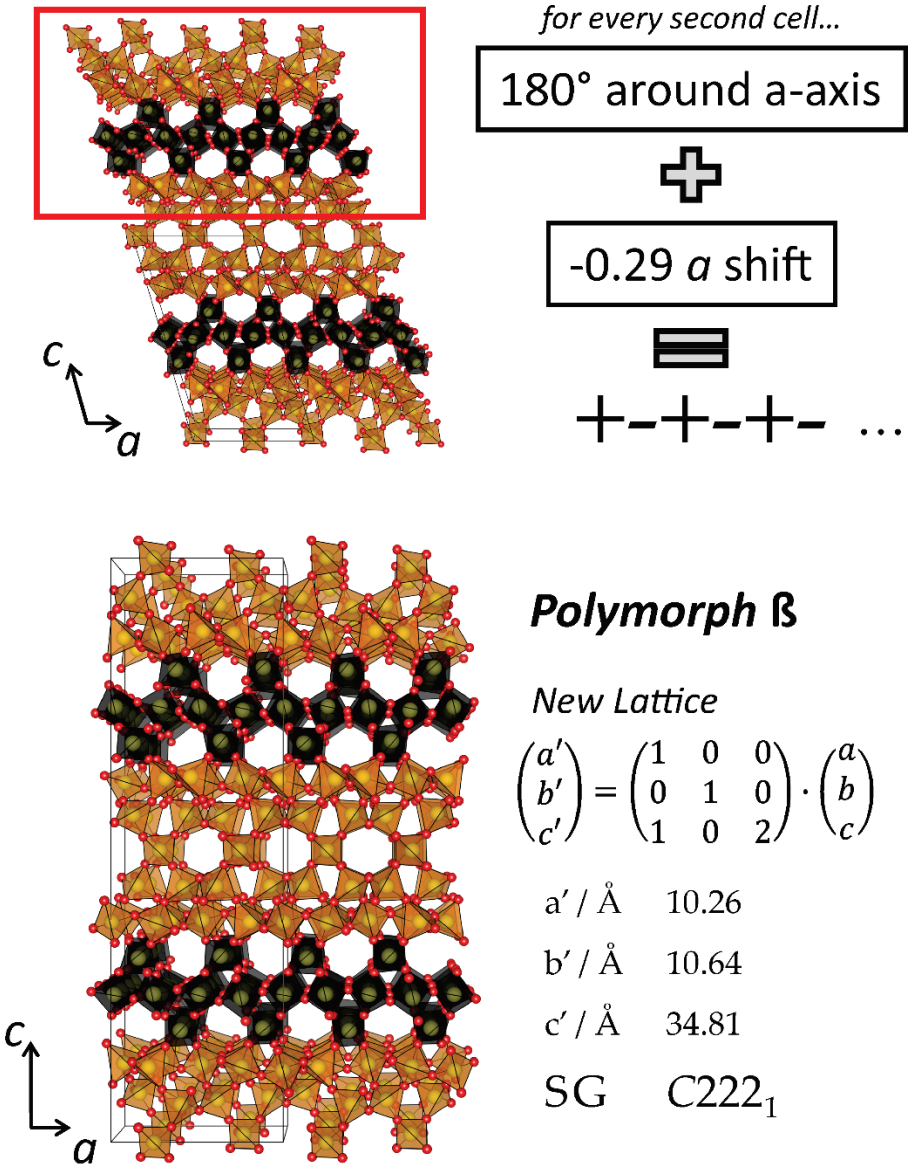

Figure S8 Construction plan to build polymorph II from polymorph I of RUB-5.

2. Disorder: Amount of RUB-6 in RUB-5

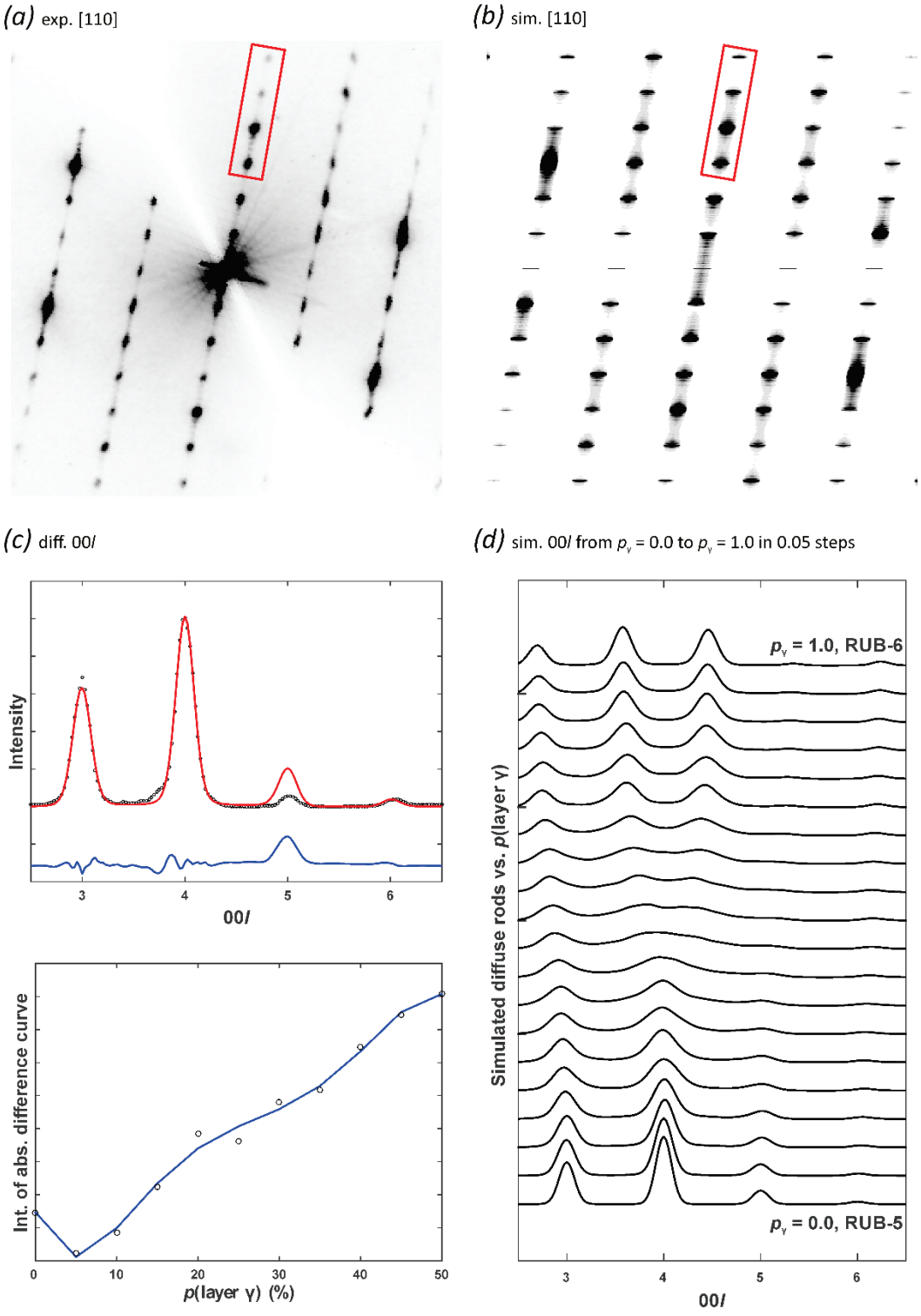

Figure S9 Experimental (a) and simulated (b) diffraction patterns of zone [110] including 00*l* reflections computed by *diffuse_extractor*. Plot of experimental line profile 00*l* (black circles) and the simulated line profile 00*l* (red) of the stacking probability p_γ = 0.05, the corresponding difference curve (blue) and plot of integrated absolute difference between simulated and experimental line profile taken along 00*l* against stacking probability of layer γ (c). Intensity profiles taken along

corresponding diffraction lines marked in (a) and (b) by red rectangles. The simulated line profiles are plotted for $p_y = 0.00$ to $p_y = 1.00$ in 0.05 steps from bottom to top (d).

3. Comparison of RUB-5 and RUB-6's diffuse scattering

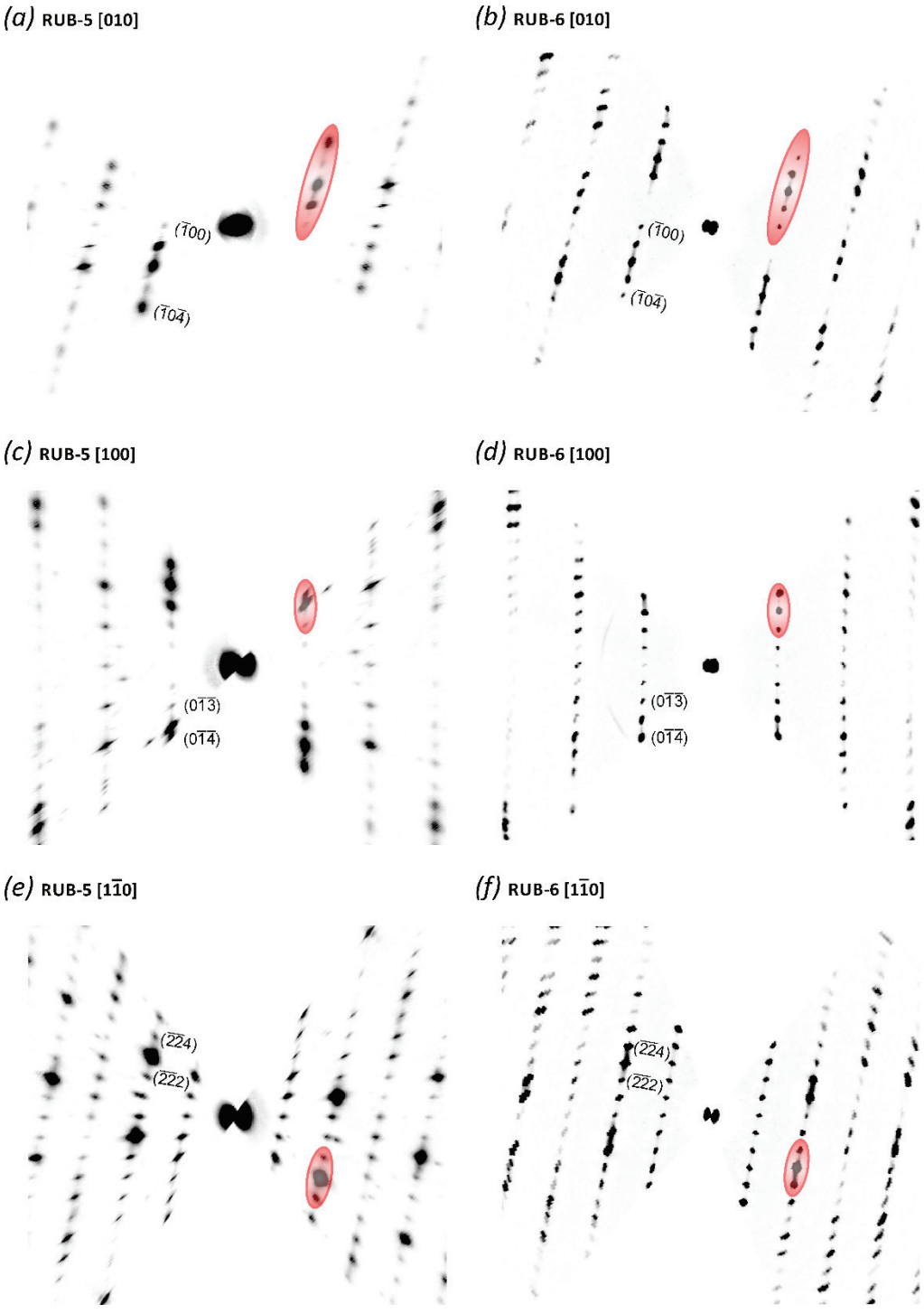

Figure S10 Experimental zones of RUB-5 and RUB-6 ADT datasets, extracted with *diffuse_extractor*, showing comparable diffuse streaks in zones [010] (a, b), [100] (c, d) and [1-10] (e, f). Selected regions are marked in red to indicate the similarity of the two compounds in reciprocal space.