Supplementary Information

Zeolite morphology and catalyst performance: Conversion of methanol to hydrocarbons over offretite

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[c]*Department of Chemistry, University of Turin, via Quarello 15, I-10135 Turin, Italy*
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Table of Contents

1. N_2 adsorption-desorption isotherms

All the N_2 sorption isotherms show a type I isotherm as expected for microporous materials.

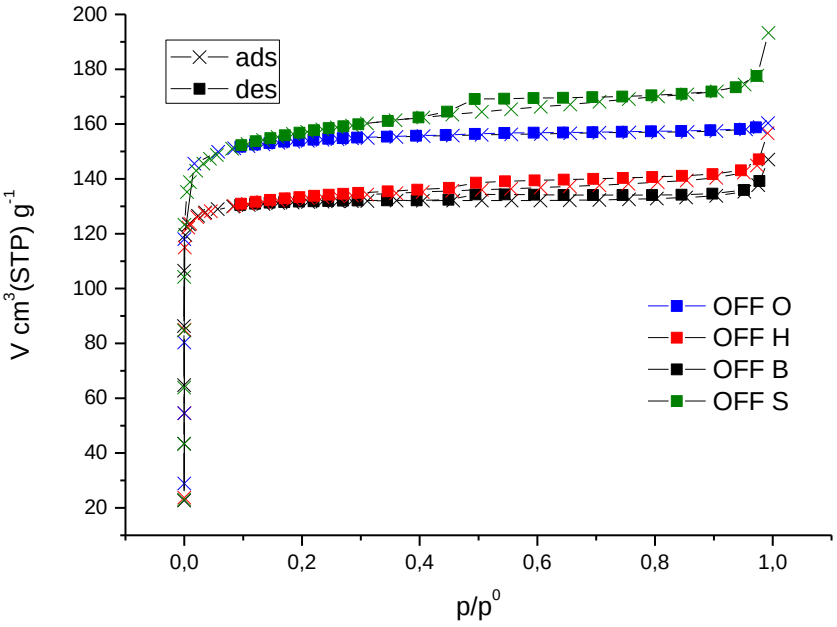

Figure 1. N_2 adsorption-desorption isotherms of offretite crystals with different shapes.

2. Material synthesis and characterization (SEM, XRD)

The offretite synthesis procedure described below allows obtaining offtetite crystals (OFF-H-b) with hexagonal morphology (Figure 2) similar to OFF-H crystals. The significant impurities of PHI phase (Figure 3) disqualified the sample for catalytic testing. However, the obtained separated crystals of offretite were suitable of SC-XRD analysis.

The precursor gel had the following batch composition: 4.8 Na_2O: 1.0 K_2O: 1.0Al_2O_3: 15.8 SiO_2: 249.5 H_2O: 1.0TMACl: 15.8 Ethanol. 4.31 g of sodium hydroxide (Sigma Aldrich, 98%) was dissolved in distilled 3.50 g of water, followed by addition of 6.94 g of aluminum isopropoxide (Sigma Aldrich, 98%). The mixture was stirred at 100 °C until dissolution. 7.22 g of distilled water was added before cooling the solution to room temperature. A separate solution was prepared in which 2.15 g of potassium hydroxide (Sigma Aldrich 87%), 2.23 g of

sodium hydroxide (Sigma-Aldrich 98%) was dissolved in 47.07 g of distilled water. 15.80 g of fumed silica (Sigma Aldrich, particle size 0.0014 µm) was added to this solution under vigorous stirring for 30 minutes. The alumina solution was introduced to the siliceous solution with further stirring for 30 minutes. 15.68 g of distilled water was added to the solution and stirred for 30 minutes. 1.88 g of tetramethylammonium chloride was introduced while stirring for 30 minutes. Finally, 12.14g of ethanol (Sigma Aldrich, 99.8%) was added as the last step of gel preparation. The precursor solution was introduced to Teflon liners which were placed in autoclaves and heated to 190 °C for 5 h. In the next step the autoclaves were submerged in cold water to return them to room temperature. The product was filtered and washed using distilled water and dried overnight at 110 °C.

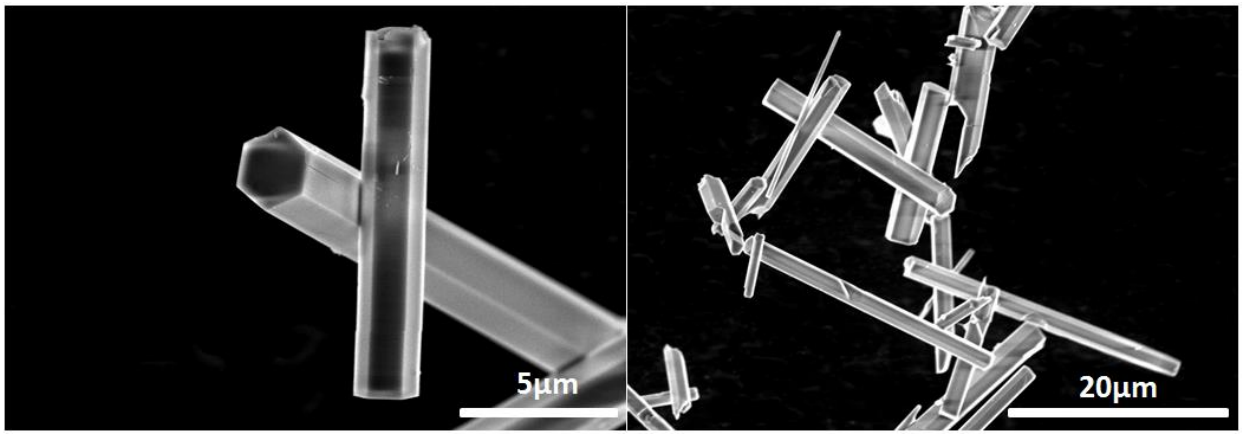

Figure 2. SEM images showing hexagonal-shaped crystals of OFF-H2

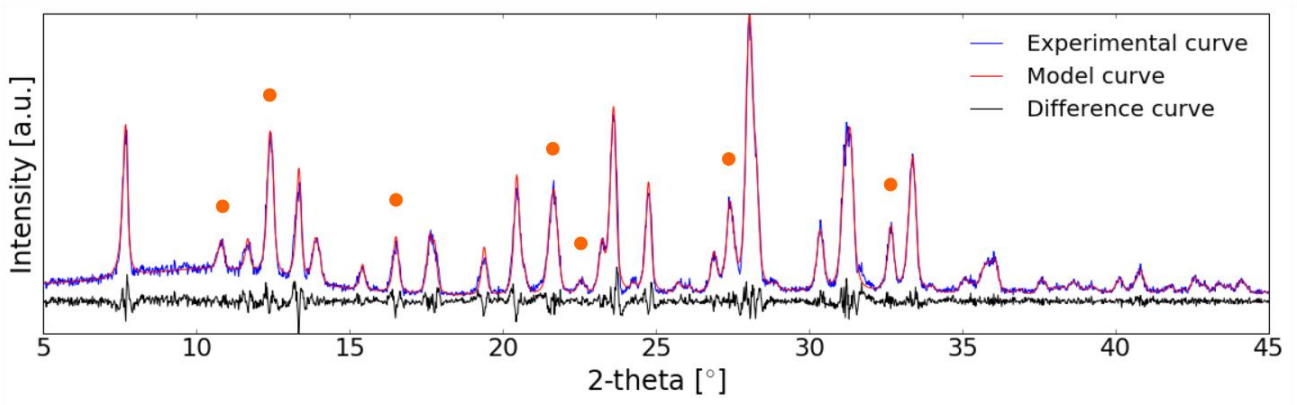

Figure 3**.** LeBail fit of experimental data of OFF-H-b performed in TOPAS. The orange dots indicate significant impurities of the PHI phase

3. Acidity characterization by FT-IT spectroscopy

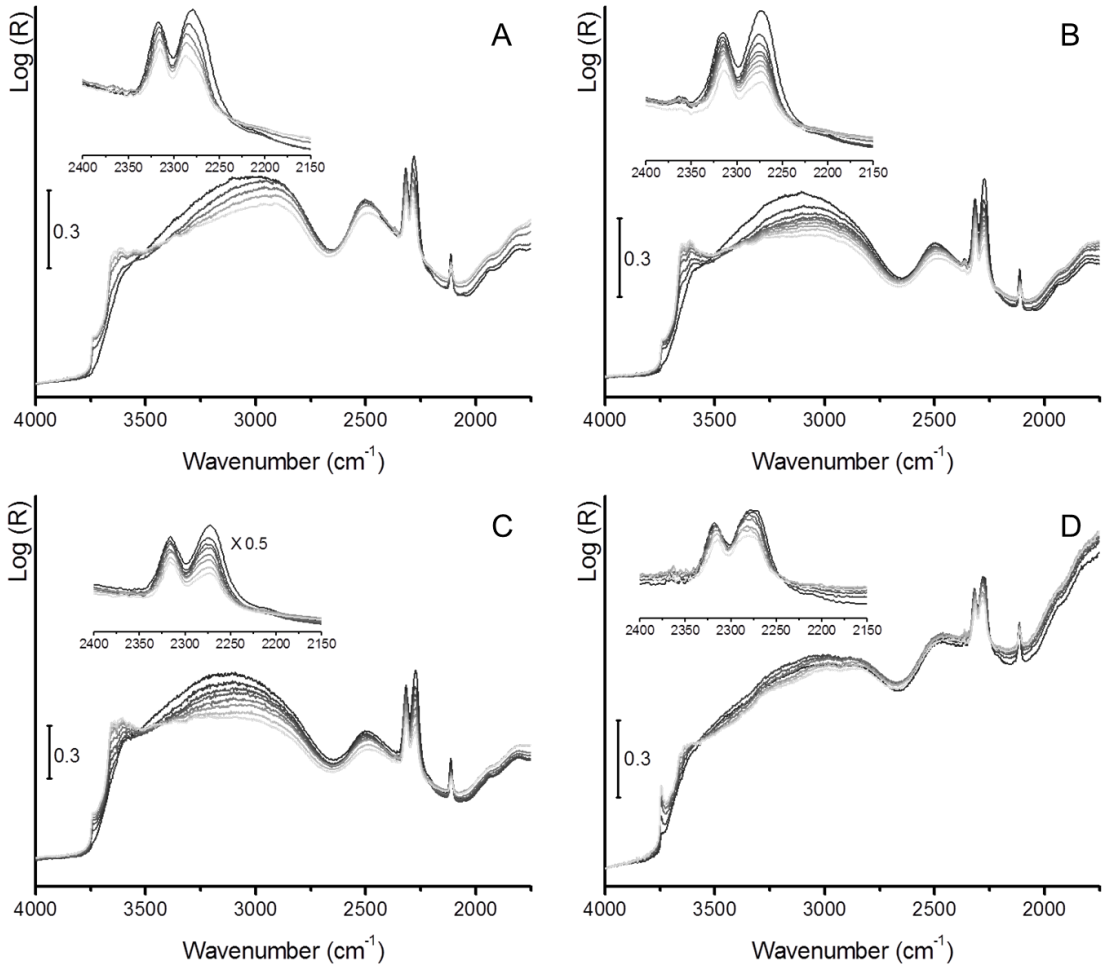

Figure 4. DRIFT spectra of CD3CN adsorbed on oval shaped offretite (OFF−O, part A), hexagonal offretite crystals (OFF−H, part B), broccoli−like shaped offretite (OFF−B, part C) and spherical offretite crystals (OFF−S, part D). Black curves represent the maximum.

Figure 4. DRIFT spectra of CD3CN adsorbed on oval shaped offretite (OFF−O, part A), hexagonal offretite crystals (OFF−H, part B), broccoli−like shaped offretite (OFF−B, part C) and spherical offretite crystals (OFF−S, part D). Black curves represent the maximumshows the IR spectra of the samples at the maximum coverage of CD_3CN (θ_{MAX}, black curve) and a selection of spectra collected along the desorption steps at 120 °C, from darkest to brightest grey. The spectra confirm the strong acidity of the samples, since only a fraction of the chemisorbed CD_3CN is removed, despite the fact that both adsorption and desorption steps were performed well above acetonitrile boiling point (80.7 °C). The larger reversibility of the components at 2272 cm^{-1}, accompanied by the faster recovery of the band assigned to OH stretching at 3740 cm^{-1}, allows us to assignee it to the CD3CN adsorbed on external SiOH groups. The more

resistant counterpart at higher frequency (2282 cm^{-1}) can be assigned to CD_3CN chemisorbed on strong Brønsted acid sites, only partially recovered upon the prolonged flux in inert. Finally, the feature centered at 2315 cm^{-1}, also quite resistant at the desorption treatment; can be associated to some extra−framework Al^{3+} Lewis center[27].

4. Adsorption isotherms of benzene

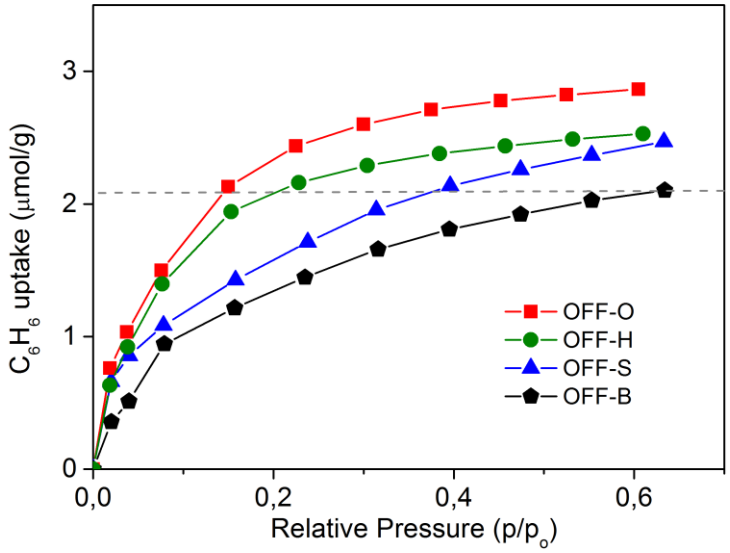

Figure 5. Experimental uptake of benzene on offretite with different morphologies at 25°C normalized by mass. The horizontal dashed line indicates total theoretical uptake.

5. Predicted and experimental crystal morphology of zeolites with CHA and MOR framework topologies

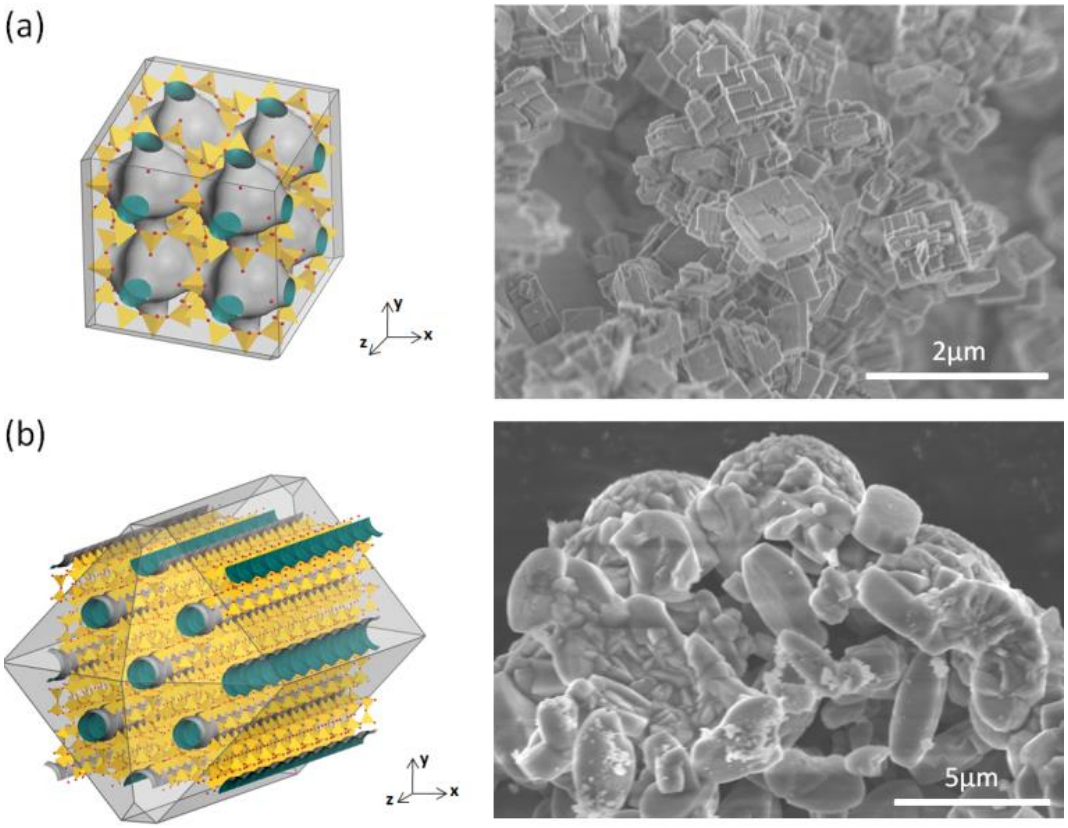

Figure 6. The visualization of (a) 3-dimensional 8MR pore system in CHA and (b) 1-dimeniosnal 12MR channels in MOR topology incorporated in simulated (BFDH method) morphology. The corresponding SEM images (on the right hand side) show the experimental crystal shape.

6. SC-XRD analysis

OFF-O crystals correspond to OFF-O-1 and OFF-O-2 labels;

OFF-H-b crystals correspond to OFF-H-b-1 and OFF-H-b2 labels

in the data below

checkCIF/PLATON report

You have not supplied any structure factors. As a result the full set of tests cannot be run.

THIS REPORT IS FOR GUIDANCE ONLY. IF USED AS PART OF A REVIEW PROCEDURE FOR PUBLICATION, IT SHOULD NOT REPLACE THE EXPERTISE OF AN EXPERIENCED CRYSTALLOGRAPHIC REFEREE.

No syntax errors found. CIF dictionary Interpreting this report

Datablock: OFF-O-1

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Bond precision:  O- C = 0.0640 A          Wavelength=3/4

Cell:              a=13.265(3)      b=13.265(3)      c=7.545(2)
                   alpha=90         beta=90          gamma=120
Temperature:       273 K

                Calculated                Reported
Volume          1149.8(7)                 1149.7(6)
Space group     P -6 m 2                  P -6 m 2
Hall group      P -6 2                    P -6 2
Moiety formula  2(O18 Si9), C5 O, 12(C), K  K0.08 O3 Si1.5, C0.42 O0.08, C
Sum formula     C17 K O37 Si18            C0.75 K0.08 O3.52 Si1.50
Mr              1340.89                   110.73
Dx,g cm-3       1.936                     1.919
Z               1                         12
Mu (mm-1)       0.802                     0.815
F000            669.0                     663.0
F000'           671.29
h,k,lmax        15,15,9                   15,15,7
Nref            869[ 467]                 694
Tmin,Tmax                                 0.494,0.745
Tmin'

Correction method= # Reported T Limits: Tmin=0.494 Tmax=0.745
AbsCorr = MULTI-SCAN

Data completeness= 1.49/0.80      Theta(max)= 26.883

R(reflections)= 0.0569( 553)      wR2(reflections)= 0.1626( 694)

S = 1.144                 Npar= 77
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The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level A

PLAT029_ALERT_3_A _diffrn_measured_fraction_theta_full value Low . 0.817 Note

Author Response: The diffractometer was single axis (synchrotron), and the crystal was weakly diffracting. Thus, low completeness was obtained.

Alert level C

PLAT041_ALERT_1_C Calc. and Reported SumFormula Strings Differ Please Check
PLAT043_ALERT_1_C Calculated and Reported Mol. Weight Differ by .. 12.13 Check
PLAT090_ALERT_3_C Poor Data / Parameter Ratio (Zmax > 18) ..... 6.06 Note
PLAT244_ALERT_4_C Low 'Solvent' Ueq as Compared to Neighbors of O00A Check
PLAT752_ALERT_4_C Angle Calc 90.00, Rep 90.00(1) ..... Senseless s.u.
C0AA -O00A -C00C 1.555 1.555 1.555 # 98 Check

Alert level G

FORMU01_ALERT_1_G There is a discrepancy between the atom counts in the
_chemical_formula_sum and _chemical_formula_moiety. This is
usually due to the moiety formula being in the wrong format.
Atom count from _chemical_formula_sum: C0.75 K.08 O3.52 Si1.5
Atom count from _chemical_formula_moiety:C1.42 K.08 O3.08 Si1.5
FORMU01_ALERT_2_G There is a discrepancy between the atom counts in the
_chemical_formula_sum and the formula from the _atom_site* data.
Atom count from _chemical_formula_sum:C0.75 K.08 O3.52 Si1.5
Atom count from the _atom_site data: C1.416666 K.0833333 O3.083333
ABSMU01_ALERT_1_G Calculation of _exptl_absorpt_correction_mu
not performed for this radiation type.
CELLZ01_ALERT_1_G Difference between formula and atom_site contents detected.
CELLZ01_ALERT_1_G ALERT: Large difference may be due to a
symmetry error - see SYMMG tests
From the CIF: _cell_formula_units_Z 12
From the CIF: _chemical_formula_sum C0.75 K0.08 O3.52 Si1.50
TEST: Compare cell contents of formula and atom_site data

atom	Z*formula	cif sites	diff
C	9.00	17.00	-8.00
K	0.96	1.00	-0.04
O	42.24	37.00	5.24
Si	18.00	18.00	0.00

PLAT003_ALERT_2_G Number of Uiso or Uij Restrained non-H Atoms ... 3 Report
PLAT004_ALERT_5_G Polymeric Structure Found with Maximum Dimension 1 Info
PLAT012_ALERT_1_G No _shelx_res_checksum found in CIF ..... Please Check
PLAT040_ALERT_1_G No H-atoms in this Carbon Containing Compound .. Please Check
PLAT042_ALERT_1_G Calc. and Reported MoietyFormula Strings Differ Please Check
PLAT045_ALERT_1_G Calculated and Reported Z Differ by a Factor ... 0.08 Check
PLAT186_ALERT_4_G The CIF-Embedded .res File Contains ISOR Records 1 Report
PLAT199_ALERT_1_G Reported _cell_measurement_temperature ..... (K) 273 Check
PLAT200_ALERT_1_G Reported _diffrn_ambient_temperature ..... (K) 273 Check
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for O004 170.9 Degree
PLAT432_ALERT_2_G Short Inter X...Y Contact Si02 .. C2 .. 3.57 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact Si02 .. C2 .. 3.57 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact O007 .. C2 .. 2.82 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact C1 .. C2 .. 2.06 Ang.

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PLAT432_ALERT_2_G Short Inter X...Y Contact  C1     ..  C2      ..       2.06 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C00B   ..  C2      ..       3.11 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C00B   ..  C2      ..       3.11 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C00B   ..  C2      ..       3.11 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C00B   ..  C2      ..       3.11 Ang.
PLAT720_ALERT_4_G Number of Unusual/Non-Standard Labels .........         13 Note
PLAT860_ALERT_3_G Number of Least-Squares Restraints ............        18 Note
PLAT870_ALERT_4_G ALERTS Related to Twinning Effects Suppressed ..         ! Info
PLAT952_ALERT_5_G Calculated (ThMax) and CIF-Reported Lmax Differ          2 Units
PLAT984_ALERT_1_G The  K-f'=     0.227 Deviates from the B&C-Value      0.217 Check
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```
   1 ALERT level A = Most likely a serious problem - resolve or explain
   0 ALERT level B = A potentially serious problem, consider carefully
   5 ALERT level C = Check. Ensure it is not caused by an omission or oversight
  29 ALERT level G = General information/check it is not something unexpected

  13 ALERT type 1 CIF construction/syntax error, inconsistent or missing data
  12 ALERT type 2 Indicator that the structure model may be wrong or deficient
   3 ALERT type 3 Indicator that the structure quality may be low
   5 ALERT type 4 Improvement, methodology, query or suggestion
   2 ALERT type 5 Informative message, check
```

It is advisable to attempt to resolve as many as possible of the alerts in all categories. Often the minor alerts point to easily fixed oversights, errors and omissions in your CIF or refinement strategy, so attention to these fine details can be worthwhile. In order to resolve some of the more serious problems it may be necessary to carry out additional measurements or structure refinements. However, the purpose of your study may justify the reported deviations and the more serious of these should normally be commented upon in the discussion or experimental section of a paper or in the "special_details" fields of the CIF. checkCIF was carefully designed to identify outliers and unusual parameters, but every test has its limitations and alerts that are not important in a particular case may appear. Conversely, the absence of alerts does not guarantee there are no aspects of the results needing attention. It is up to the individual to critically assess their own results and, if necessary, seek expert advice.

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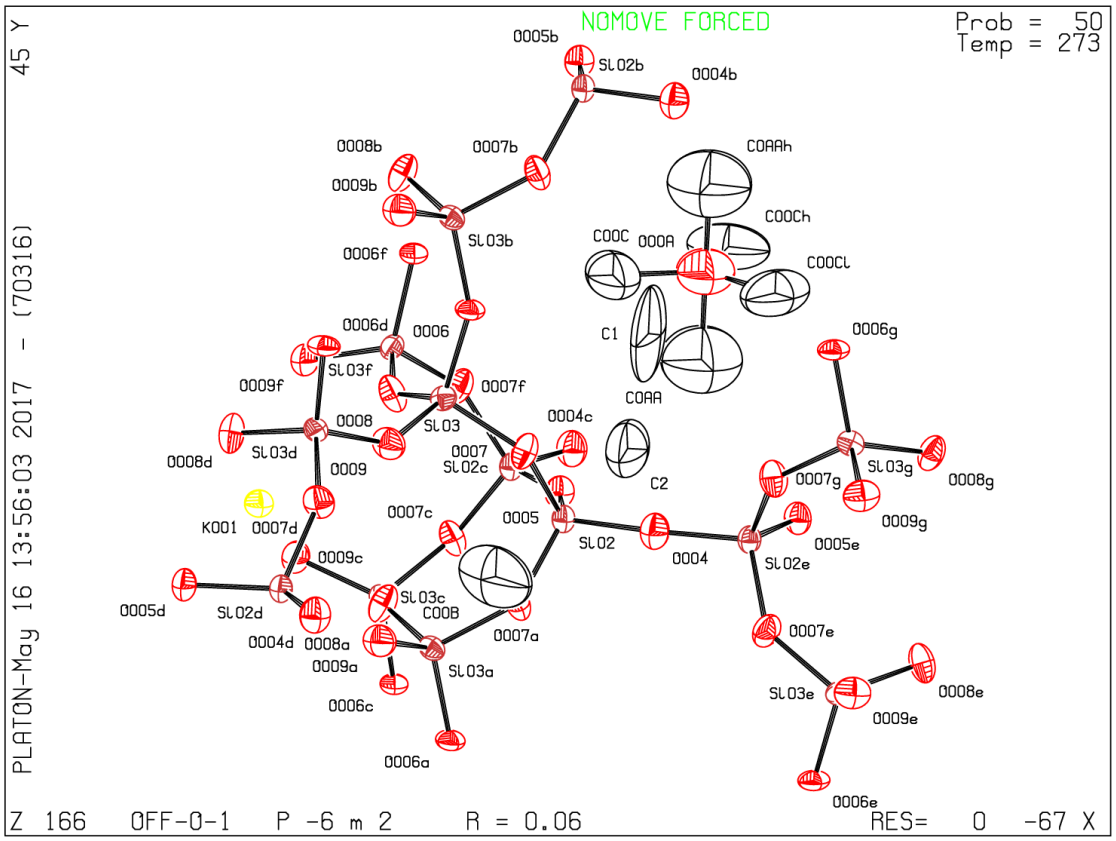
PLATON version of 27/03/2017; check.def file version of 24/03/2017

Datablock OFF-O-1 - ellipsoid plot



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You have not supplied any structure factors. As a result the full set of tests cannot be run.

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No syntax errors found. CIF dictionary Interpreting this report

Datablock: OFF-O-2

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Cell:             a=13.2558(3)      b=13.2558(3)      c=7.5358(2)
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Temperature:      273 K
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Volume	1146.76(7)	1146.76(6)
Space group	P -6 m 2	P -6 m 2
Hall group	P -6 2	P -6 2
Moiety formula	2(O18 Si9), C5 O, 3(O), 9(C), K	K1 O36 Si18, 2(C2.5 O0.5), 9(C), 3(O)
Sum formula	C14 K O40 Si18	C14 K O40 Si18
Mr	1352.86	1352.86
Dx,g cm-3	1.959	1.959
Z	1	1
Mu (mm-1)	0.810	0.817
F000	675.0	675.0
F000’	677.32	
h,k,lmax	18,18,10	18,18,9
Nref	1245[664]	1065
Tmin,Tmax		0.650,0.746
Tmin’		

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R(reflections)= 0.0431( 1009)     wR2(reflections)= 0.1322( 1065)

S = 1.112                 Npar= 77
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The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level B

PLAT306_ALERT_2_B Isolated Oxygen Atom (H-atoms Missing ?) ..... O1 Check

Author Response: Residual disordered electron density in the pores was modelled using dummy O and C atoms.

Alert level C

STRVA01_ALERT_4_C Flack parameter is too small
From the CIF: _refine_ls_abs_structure_Flack -0.240
From the CIF: _refine_ls_abs_structure_Flack_su 0.100
PLAT244_ALERT_4_C Low 'Solvent' Ueq as Compared to Neighbors of O00A Check
PLAT752_ALERT_4_C Angle Calc 90.00, Rep 90.00(1) ..... Senseless s.u.
C4 -O00A -C0AA 1.555 1.555 1.555 # 99 Check

Alert level G

ABSMU01_ALERT_1_G Calculation of _exptl_absorpt_correction_mu
not performed for this radiation type.
PLAT004_ALERT_5_G Polymeric Structure Found with Maximum Dimension 1 Info
PLAT012_ALERT_1_G No _shelx_res_checksum found in CIF ..... Please Check
PLAT019_ALERT_1_G _diffrn_measured_fraction_theta_full/*_max < 1.0 0.973 Report
PLAT040_ALERT_1_G No H-atoms in this Carbon Containing Compound .. Please Check
PLAT042_ALERT_1_G Calc. and Reported MoietyFormula Strings Differ Please Check
PLAT199_ALERT_1_G Reported _cell_measurement_temperature ..... (K) 273 Check
PLAT200_ALERT_1_G Reported _diffrn_ambient_temperature ..... (K) 273 Check
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for O005 171.0 Degree
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for O007 139.5 Degree
PLAT432_ALERT_2_G Short Inter X...Y Contact Si03 .. C2 .. 3.52 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact Si03 .. C2 .. 3.52 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact O009 .. C2 .. 2.77 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact C2 .. C3 .. 2.12 Ang.
PLAT720_ALERT_4_G Number of Unusual/Non-Standard Labels ..... 11 Note
PLAT792_ALERT_1_G The Model has Chirality at Si03 (Polar SPGR) S Verify
PLAT870_ALERT_4_G ALERTS Related to Twinning Effects Suppressed .. ! Info
PLAT984_ALERT_1_G The K-f'= 0.227 Deviates from the B&C-Value 0.217 Check

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0 **ALERT level A** = Most likely a serious problem - resolve or explain
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9 ALERT type 1 CIF construction/syntax error, inconsistent or missing data
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0 ALERT type 3 Indicator that the structure quality may be low
5 ALERT type 4 Improvement, methodology, query or suggestion
1 ALERT type 5 Informative message, check

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Publication of your CIF in IUCr journals

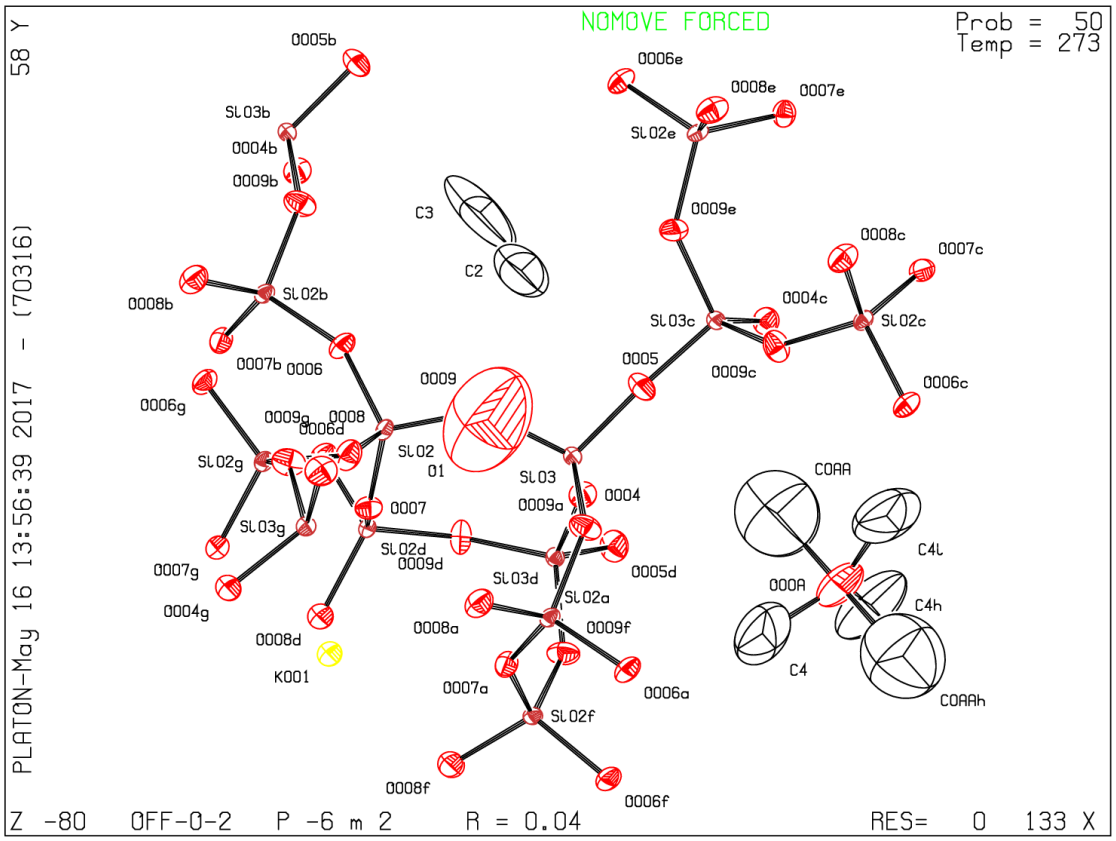
A basic structural check has been run on your CIF. These basic checks will be run on all CIFs submitted for publication in IUCr journals (*Acta Crystallographica, Journal of Applied Crystallography, Journal of Synchrotron Radiation*); however, if you intend to submit to *Acta Crystallographica Section C* or *E* or *IUCrData*, you should make sure that full publication checks are run on the final version of your CIF prior to submission.

Publication of your CIF in other journals

Please refer to the *Notes for Authors* of the relevant journal for any special instructions relating to CIF submission.

PLATON version of 27/03/2017; check.def file version of 24/03/2017

Datablock OFF-O-2 - ellipsoid plot



checkCIF/PLATON report

You have not supplied any structure factors. As a result the full set of tests cannot be run.

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No syntax errors found. CIF dictionary Interpreting this report

Datablock: OFF-H-b-1

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Bond precision:  O- C = 0.0300 A          Wavelength=3/4

Cell:             a=13.2511(3)      b=13.2511(3)      c=7.5361(2)
                  alpha=90          beta=90           gamma=120
Temperature:      273 K
```

	Calculated	Reported
Volume	1145.99(7)	1145.99(6)
Space group	P -6 m 2	P -6 m 2
Hall group	P -6 2	P -6 2
Moiety formula	2(O18 Si9), C5 O, 3(O), 9(C), K	K1 O36 Si18, 2(C2.5 O0.5), 3(O), 9(C)
Sum formula	C14 K O40 Si18	C14 K O40 Si18
Mr	1352.86	1352.91
Dx,g cm-3	1.960	1.960
Z	1	1
Mu (mm-1)	0.811	0.817
F000	675.0	675.0
F000’	677.32	
h,k,lmax	17,17,10	17,17,10
Nref	1194[637]	1185
Tmin,Tmax		0.654,0.746
Tmin’		

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Correction method= # Reported T Limits: Tmin=0.654 Tmax=0.746
AbsCorr = MULTI-SCAN

Data completeness= 1.86/0.99      Theta(max)= 30.404

R(reflections)= 0.0465( 1038)     wR2(reflections)= 0.1405( 1185)

S = 1.058                 Npar= 77
```

The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level B

```
CHEMW03_ALERT_2_B WARNING: The ratio of given/expected molecular weight as
            calculated from the _atom_site* data lies outside
            the range 0.95 <> 1.05
           From the CIF: _cell_formula_units_Z                    1
           From the CIF: _chemical_formula_weight          1352.91
           TEST: Calculate formula weight from _atom_site_*
           atom     mass    num     sum
           C        12.01   17.00  204.20
           K        39.10    1.00   39.10
           O        16.00   43.00  687.98
           Si       28.09   18.00  505.57
           Calculated formula weight             1436.85
PLAT306_ALERT_2_B Isolated Oxygen Atom (H-atoms Missing ?) .......       O00B Check
```

Author Response: Residual disordered electron density in the pores was modelled using dummy O and C atoms.

Alert level C

```
PLAT244_ALERT_4_C Low    'Solvent' Ueq as Compared to Neighbors of       O00A Check
PLAT752_ALERT_4_C Angle   Calc       90.00, Rep       90.00 ......  Senseless s.u.
             C4     -O00A   -C1         1.555   1.555   1.555     #   96 Check
```

Alert level G

```
FORMU01_ALERT_2_G  There is a discrepancy between the atom counts in the
            _chemical_formula_sum and the formula from the _atom_site* data.
            Atom count from _chemical_formula_sum:C14 K1 O40 Si18
            Atom count from the _atom_site data:  C17. K1. O43.00172 Si18.
ABSMU01_ALERT_1_G  Calculation of _exptl_absorpt_correction_mu
               not performed for this radiation type.
CELLZ01_ALERT_1_G Difference between formula and atom_site contents detected.
CELLZ01_ALERT_1_G ALERT: Large difference may be due to a
            symmetry error - see SYMMG tests
           From the CIF: _cell_formula_units_Z    1
           From the CIF: _chemical_formula_sum  C14 K O40 Si18
           TEST: Compare cell contents of formula and atom_site data

           atom    Z*formula  cif sites diff
           C         14.00     17.00   -3.00
           K          1.00      1.00    0.00
           O         40.00     43.00   -3.00
           Si        18.00     18.00    0.00
PLAT004_ALERT_5_G Polymeric Structure Found with Maximum Dimension          1 Info
PLAT012_ALERT_1_G No     _shelx_res_checksum found in CIF ........     Please Check
PLAT040_ALERT_1_G No H-atoms in this Carbon Containing Compound ..     Please Check
PLAT042_ALERT_1_G Calc. and Reported MoietyFormula Strings  Differ     Please Check
PLAT072_ALERT_2_G SHELXL First  Parameter in WGHT  Unusually Large       0.10 Report
PLAT199_ALERT_1_G Reported _cell_measurement_temperature ..... (K)        273 Check
PLAT200_ALERT_1_G Reported   _diffrn_ambient_temperature ..... (K)        273 Check
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for  O005        171.2 Degree
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for  O006        140.0 Degree
```

```
PLAT432_ALERT_2_G Short Inter X...Y Contact  Si03   ..  C3      ..       3.53 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  Si03   ..  C3      ..       3.53 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  O008   ..  C3      ..       2.78 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C2     ..  C2      ..       0.01 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C2     ..  C3      ..       2.09 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact  C2     ..  C3      ..       2.09 Ang.
PLAT720_ALERT_4_G Number of Unusual/Non-Standard Labels .........         11 Note
PLAT870_ALERT_4_G ALERTS Related to Twinning Effects Suppressed ..          ! Info
PLAT933_ALERT_2_G Number of OMIT Records in Embedded .res File ...          2 Note
PLAT984_ALERT_1_G The  K-f'=     0.227 Deviates from the B&C-Value      0.217 Check
```

```
   0 ALERT level A = Most likely a serious problem - resolve or explain
   2 ALERT level B = A potentially serious problem, consider carefully
   2 ALERT level C = Check. Ensure it is not caused by an omission or oversight
  23 ALERT level G = General information/check it is not something unexpected

   9 ALERT type 1 CIF construction/syntax error, inconsistent or missing data
  13 ALERT type 2 Indicator that the structure model may be wrong or deficient
   0 ALERT type 3 Indicator that the structure quality may be low
   4 ALERT type 4 Improvement, methodology, query or suggestion
   1 ALERT type 5 Informative message, check
```

It is advisable to attempt to resolve as many as possible of the alerts in all categories. Often the minor alerts point to easily fixed oversights, errors and omissions in your CIF or refinement strategy, so attention to these fine details can be worthwhile. In order to resolve some of the more serious problems it may be necessary to carry out additional measurements or structure refinements. However, the purpose of your study may justify the reported deviations and the more serious of these should normally be commented upon in the discussion or experimental section of a paper or in the "special_details" fields of the CIF. checkCIF was carefully designed to identify outliers and unusual parameters, but every test has its limitations and alerts that are not important in a particular case may appear. Conversely, the absence of alerts does not guarantee there are no aspects of the results needing attention. It is up to the individual to critically assess their own results and, if necessary, seek expert advice.

Publication of your CIF in IUCr journals

A basic structural check has been run on your CIF. These basic checks will be run on all CIFs submitted for publication in IUCr journals (*Acta Crystallographica*, *Journal of Applied Crystallography*, *Journal of Synchrotron Radiation*); however, if you intend to submit to *Acta Crystallographica Section C* or *E* or *IUCrData*, you should make sure that full publication checks are run on the final version of your CIF prior to submission.

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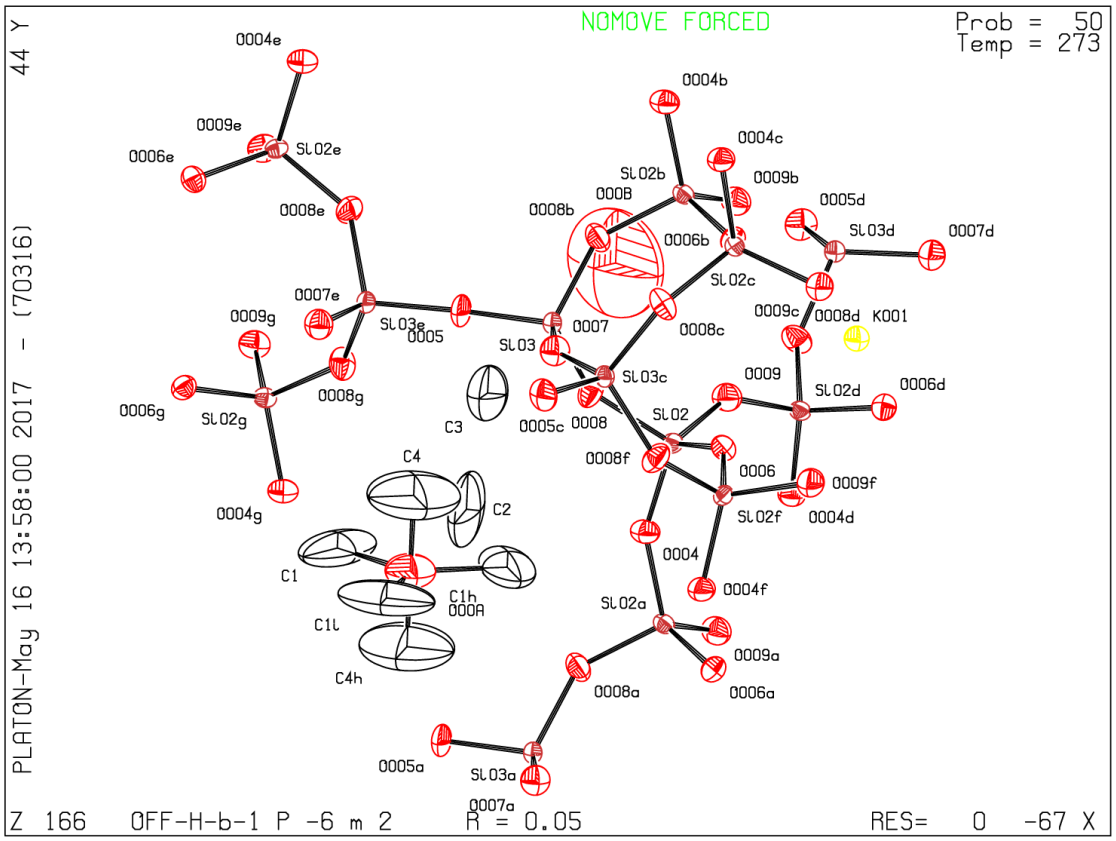
PLATON version of 27/03/2017; check.def file version of 24/03/2017

Datablock OFF-H-b-1 - ellipsoid plot



checkCIF/PLATON report

You have not supplied any structure factors. As a result the full set of tests cannot be run.

THIS REPORT IS FOR GUIDANCE ONLY. IF USED AS PART OF A REVIEW PROCEDURE FOR PUBLICATION, IT SHOULD NOT REPLACE THE EXPERTISE OF AN EXPERIENCED CRYSTALLOGRAPHIC REFEREE.

No syntax errors found. CIF dictionary Interpreting this report

Datablock: OFF-H-b-2

```
Bond precision:  O- C = 0.0260 A          Wavelength=0.71073

Cell:            a=13.2269(16)     b=13.2269(16)     c=7.5381(10)
                 alpha=90          beta=90           gamma=120
Temperature:     273 K
```

	Calculated	Reported
Volume	1142.1(4)	1142.1(3)
Space group	P -6 m 2	P -6 m 2
Hall group	P -6 2	P -6 2
Moiety formula	2(O18 Si9), C5 O, 3(O), 9(C), K	K1 O36 Si18, 2(C2.5 O0.5), 3(O), 9(C)
Sum formula	C14 K O40 Si18	C14 K O40 Si18
Mr	1352.86	1352.86
Dx,g cm-3	1.967	1.967
Z	1	1
Mu (mm-1)	0.708	0.708
F000	675.0	675.0
F000’	677.06	
h,k,lmax	17,17,10	17,17,10
Nref	1196[638]	1180
Tmin,Tmax	0.996,0.996	0.609,0.746
Tmin’	0.993	

```
Correction method= # Reported T Limits: Tmin=0.609 Tmax=0.746
AbsCorr = MULTI-SCAN

Data completeness= 1.85/0.99      Theta(max)= 28.711

R(reflections)= 0.0358( 1156)     wR2(reflections)= 0.1050( 1180)

S = 1.166                 Npar= 77
```

The following ALERTS were generated. Each ALERT has the format
test-name_ALERT_alert-type_alert-level.
Click on the hyperlinks for more details of the test.

Alert level B

PLAT306_ALERT_2_B Isolated Oxygen Atom (H-atoms Missing ?) ..... O00B Check

Author Response: Residual disordered electron density in the pores was modelled using dummy O and C atoms.

Alert level C

PLAT094_ALERT_2_C Ratio of Maximum / Minimum Residual Density .... 2.06 Report
PLAT244_ALERT_4_C Low 'Solvent' Ueq as Compared to Neighbors of O00A Check
PLAT752_ALERT_4_C Angle Calc 90.00, Rep 90.00(2) ..... Senseless s.u.
C1 -O00A -C00C 1.555 1.555 1.555 # 99 Check

Alert level G

PLAT004_ALERT_5_G Polymeric Structure Found with Maximum Dimension 1 Info
PLAT012_ALERT_1_G No _shelx_res_checksum found in CIF ..... Please Check
PLAT019_ALERT_1_G _diffrn_measured_fraction_theta_full/*_max < 1.0 0.996 Report
PLAT040_ALERT_1_G No H-atoms in this Carbon Containing Compound .. Please Check
PLAT042_ALERT_1_G Calc. and Reported MoietyFormula Strings Differ Please Check
PLAT199_ALERT_1_G Reported _cell_measurement_temperature ..... (K) 273 Check
PLAT200_ALERT_1_G Reported _diffrn_ambient_temperature ..... (K) 273 Check
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for O005 139.7 Degree
PLAT396_ALERT_2_G Deviating Si-O-Si Angle from 150 Deg for O006 171.3 Degree
PLAT432_ALERT_2_G Short Inter X...Y Contact Si03 .. C2 .. 3.53 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact Si03 .. C2 .. 3.53 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact O009 .. C2 .. 2.78 Ang.
PLAT432_ALERT_2_G Short Inter X...Y Contact C2 .. C0AA .. 2.15 Ang.
PLAT720_ALERT_4_G Number of Unusual/Non-Standard Labels ..... 13 Note
PLAT870_ALERT_4_G ALERTS Related to Twinning Effects Suppressed .. ! Info

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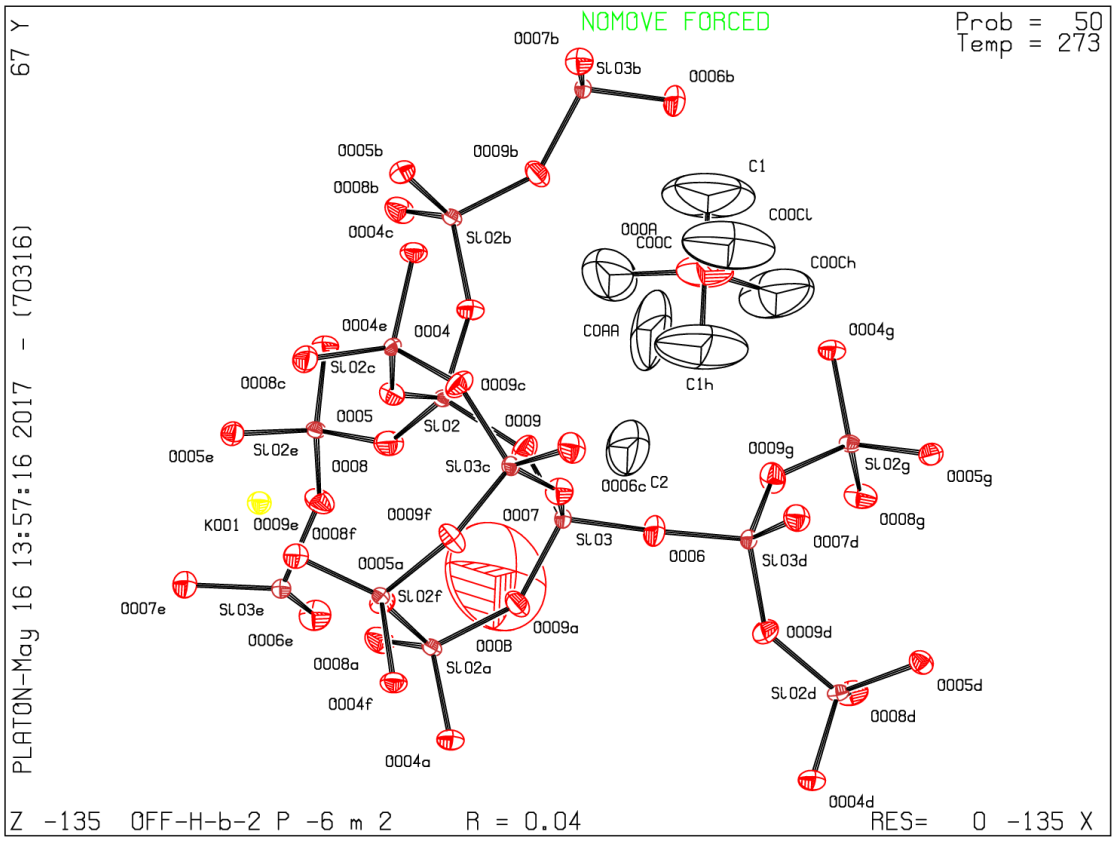
PLATON version of 27/03/2017; check.def file version of 24/03/2017

Datablock OFF-H-b-2 - ellipsoid plot



7. Summary of literature data on catalyst testing

Table S.1. Summary of literature data on catalyst testing. Reference numbering refers to the numbering used in the main manuscript.

Catalyst	Conditions	Products	Lifetime	Acidity	Morphology	Ref.
H-Offretite	T = 377 °C. WHSV = 10 h^{-1}.	No information provided.	Ca. 1 h. Maximum initial conversion was 80 %	Si/Al = 4 in catalyst.	No information provided.	6
H-Offretite (prepared with TMA).	T = 244 °C (and lower). WHSV = 1.5 h^{-1}.	Mostly DME formation. Only C_1-C_3 reported.	Ca. 3 h at 244 °C. Maximum 15 % conversion to hydrocarbons reached.	Si/Al = 4.5 in synthesis gel.	No direct information provided. Literature reference indicates oval agglomerates, about 0.3 x 1.5 µm in size, AR=5	7
Zeolite T (OFF – ERI intergrowth; inorganic synthesis)	T = 300 or 400 °C Residence time = 2.5 g_{cat} s cm^{-3}	Rich in C_2-C_4 aliphatics (71.9 wt% at 400 °C) Low selectivity to branched products. Minor amounts of aromatics.	Ca. 8 h at 400 °C. Maximum 90 % conversion to hydrocarbons reached.	Si/Al = 3.3 in catalyst.	No direct information provided. Literature reference indicates rod-shaped, 3-4 × 0.5-0.6 µm.	8
Zeolite T (OFF – ERI intergrowth)	T = 400 °C. WHSV = 1 h^{-1}.	Ethene and propene (max 75 wt%) and C_1-C_4 alkanes.	Ranging from ca. 4 h (43 % cation exchange) to 1.5 h (58 % cation	Si/Al = 3.4 in parent material. Degree of cation exchange ranging from 43 to 84 %.	No information provided.	9

			exchange, amorphous material present).	Characterization with FT-IR.		
H-Offretite (prepared with TMA, small ERI content present).	Reaction carried out in sealed NMR rotor and sequential heating to 150 – 300 °C for 10 min at each T.	For H-OFF, a range of aliphatic hydrocarbon products and also long-chain hydrocarbons trapped inside the structure.	No information provided.	Same catalysts as in refs. 11 and 12.		10
Series of OFF-ERI intergrowths with decreasing content of ERI prepared with various SDAs. TMA-offretite, CC-offretite, and BTMA-offretite have the lowest number of channel blockings due to ERI intergrowths.	T = 425 °C. LHSV = 0.5 (no units provided).	TMA-offretite: Sum of methane, ethene, and propene is maximum 67.4 C%. C_5 is 1.8 C% BTMA-offretite: Sum of methane, ethene, and propene is maximum 53.1 C%. C_5 is 0.7 C% CC-offretite: Sum of methane, ethene, and propene is maximum 58.2 C%. C_5 is 2.0 C%	Initial conversion ca. 100 % for all catalysts. Lifetime ranges from 2 to 4 h (estimated from Figure 5 in ref. 11).	TMA-offretite: Si/Al = 3.9 from elemental analysis; 5.3 from Si NMR after ion ex. and treatment at 600 °C. BTMA-offretite: Si/Al = 5.8 from elemental analysis; 9.2 from Si NMR after ion ex. and treatment at 600 °C. CC-offretite: Si/Al = 4.1 from elemental analysis; 4.0 from Si NMR after ion ex. and treatment at 500 °C.	TMA-offretite: Irregular, ca. 0.2-0.5 × 0.5-1 µm. BTMA-offretite: Micron sized hexagonal rods (scale in SEM image hard to discern). CC-offretite: Micron sized rods, ca. 1 × 5-10 µm.	11,12