

## Supporting Information

### **A Simple Matrine Derivative for the Facile Syntheses of Mesoporous Zeolites ITQ-37 and ITQ-43**

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**Synthesis of the N-methylmatrinium (MeMAS):** In a typical synthesis 24.8 g (0.1 mol) matrine and 5.7 g (0.15 mol) lithium tetrahydroaluminate were dissolved in 200 mL anhydrous tetrahydrofuran in a 500 mL round-bottomed flask. The mixture was refluxed for 10 h and then cooled to room temperature. The excess of  $\text{LiAlH}_4$  was decomposed with water. The solution was filtered and evaporated to get the reduction product of matrine (I). In the second step reaction, 23.4 g (0.1 mol) (I) was dissolved in 200 mL acetone and 28.4 g (0.2 mol)  $\text{CH}_3\text{I}$  was slowly added, the solution was refluxed for 10 h, cooled. The solid was obtained by filtration and washed with diethyl ether. The product was exchanged to the hydroxide form with an anionic exchange resin in a polyethylene bottle overnight.

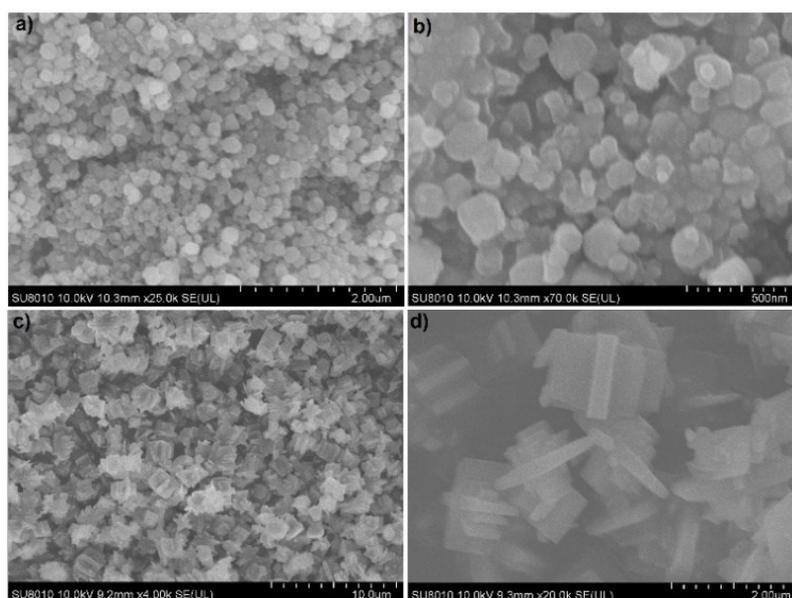
**Synthesis of MAS-ITQ-37.** A typical synthesis gel was prepared by dissolving 69.7 mg (0.67 mmol) of germanium oxide in 1.0 g (0.60 mmol) of a solution of MASOH with a concentration of 0.60 mol  $\text{OH}^-/\text{kg}$ . Then, 278 mg (1.33 mmol) of tetraethylorthosilicate (TEOS) were added and hydrolyzed under stirring at room temperature until the total hydrolysis of TEOS, and 222 mg (0.60 mmol) of  $\text{NH}_4\text{F}$  (10%) were added. The appropriate amount of water was allowed to evaporate under a stream of air. The final gel with a molar composition of  $\text{Si}/\text{Ge}=2$ ,  $\text{SDA}/\text{T}=0.3$ ,  $\text{NH}_4\text{F}/\text{T}=0.3$ ,  $\text{H}_2\text{O}/\text{T}=1.5$  ( $\text{T}=\text{Si}+\text{Ge}$ ) was transferred into Teflon-lined steel autoclaves and kept at  $160^\circ\text{C}$  for 2 days. After cooled to room temperature, the final powder was recovered by filtration, washed with distilled water and acetone, dried at  $60^\circ\text{C}$  overnight.

**Synthesis of MAS-ITQ-43.** Except for the addition of a small amount of aluminium isopropoxide (10.2 mg, 0.05 mmol), and a final gel with a molar composition of  $\text{Si}/\text{Ge}=2$ ,  $\text{Al}/\text{T}=0.025$ ,  $\text{SDA}/\text{T}=0.3$ ,  $\text{NH}_4\text{F}/\text{T}=0.3$ ,  $\text{H}_2\text{O}/\text{T}=3$ , the synthesis process and crystallization condition of MAS-ITQ-43 were the same as that of the above mentioned synthesis of MAS-ITQ-37.

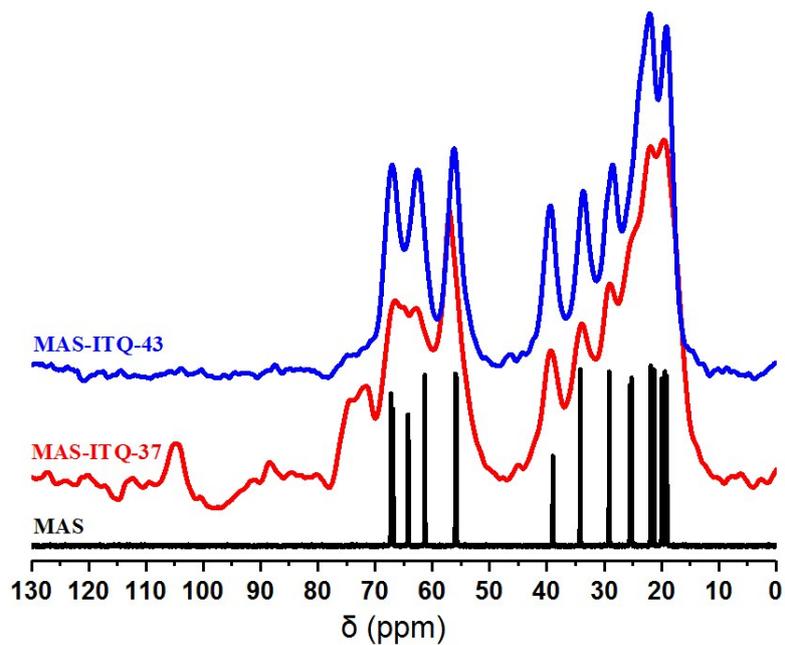
### Characteristics :

Liquid NMR spectra were recorded with a 500 MHz Spectrometer. The XRD patterns were collected on a Rigaku SmartLab diffractometer with Cu K $\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ). The scanning electron microscopy (SEM) images were taken on an Ultra-high Resolution FE-SEM SU8010 electron microscope. Thermogravimetric analysis was performed on a Q500 TGA unit in air with a heating rate of  $10^\circ\text{C}/\text{min}$  from room temperature to  $800^\circ\text{C}$ . The  $\text{N}_2$  adsorption-desorption isotherms of MAS-ITQ-37 and MAS-ITQ-43 at 77 K were measured with a Micrometrics ASAP 2020 Plus instrument. Solid-state MAS-NMR spectra of  $^{13}\text{C}$ 、 $^{19}\text{F}$ 、 $^{27}\text{Al}$  were measured at Bruker Avance III 400 WB.

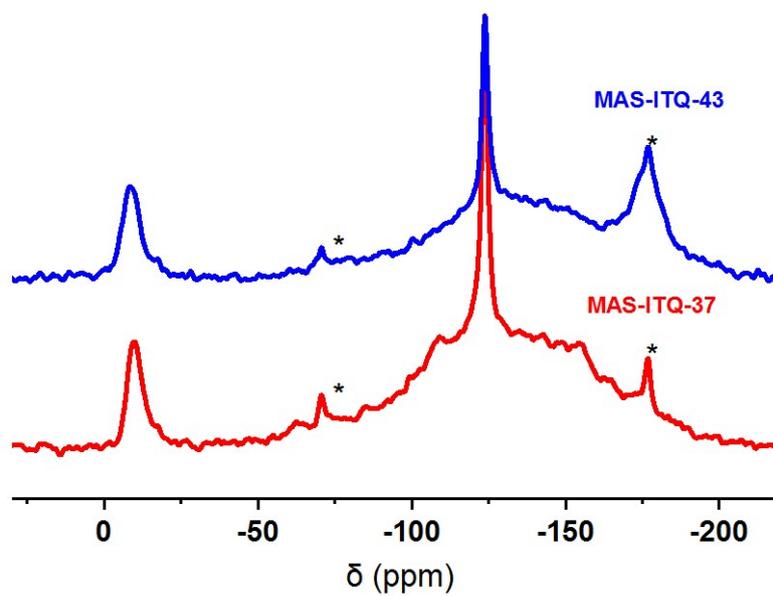
**Crystal structure determination.** Single-crystal X-ray diffraction measurement was performed on a Bruker AXS SMART APEX-II diffractometer for MeMAS in its iodide with graphite monochromated Mo K $\alpha$  ( $\lambda = 0.71073 \text{ \AA}$ ) radiation at 293 K. Data processing was completed with the SAINT program.<sup>S1</sup> The structure of MeMAT was solved by the direct method and refined on  $F^2$  by full-matrix least-squares with the SHELX-97 program.<sup>S2</sup> A summary of the crystallographic data is given in Table S2.



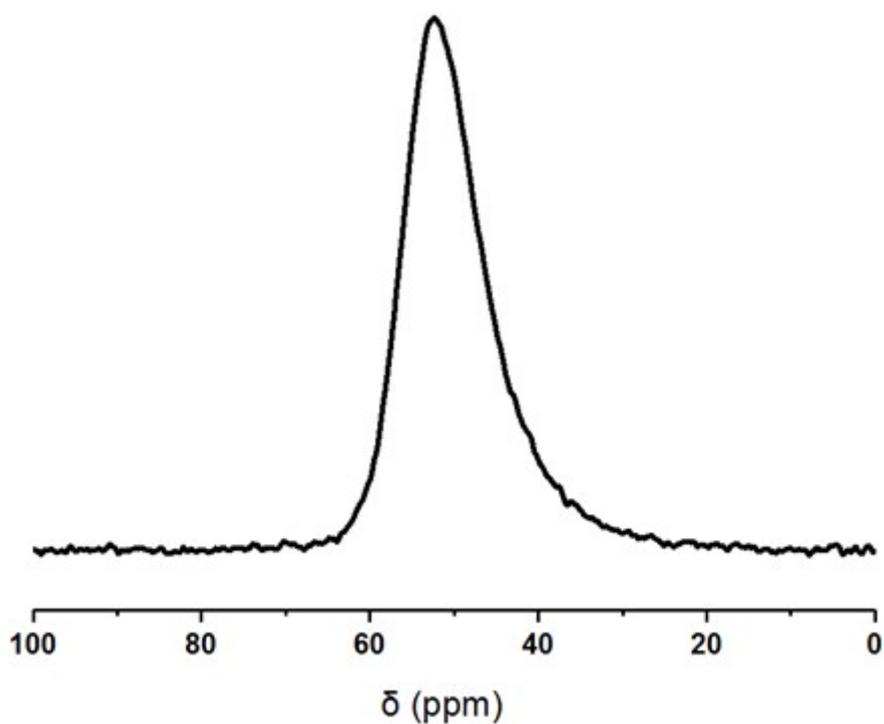
**Figure S1.** SEM images of the as-synthesized MAS-ITQ-37 (a, b) and MAS-ITQ-43 (c, d).



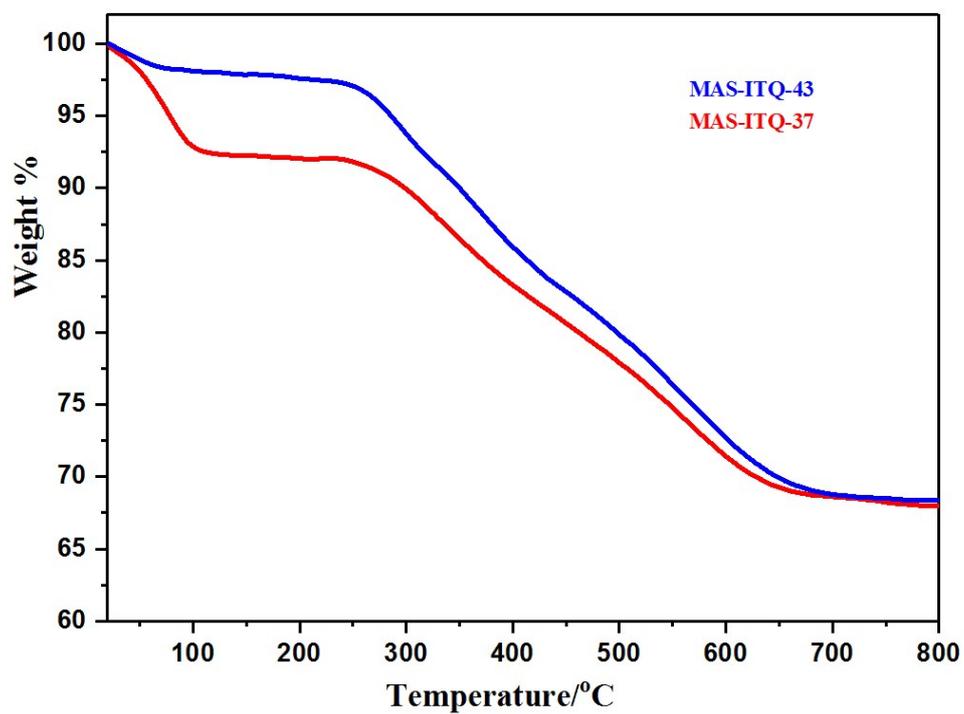
**Figure S2.**  $^{13}\text{C}$  liquid NMR spectrum of OSDA in  $\text{D}_2\text{O}$  and  $^{13}\text{C}$  solid-state MAS NMR spectra of the as-synthesized MAS-ITQ-37 and MAS-ITQ-43.



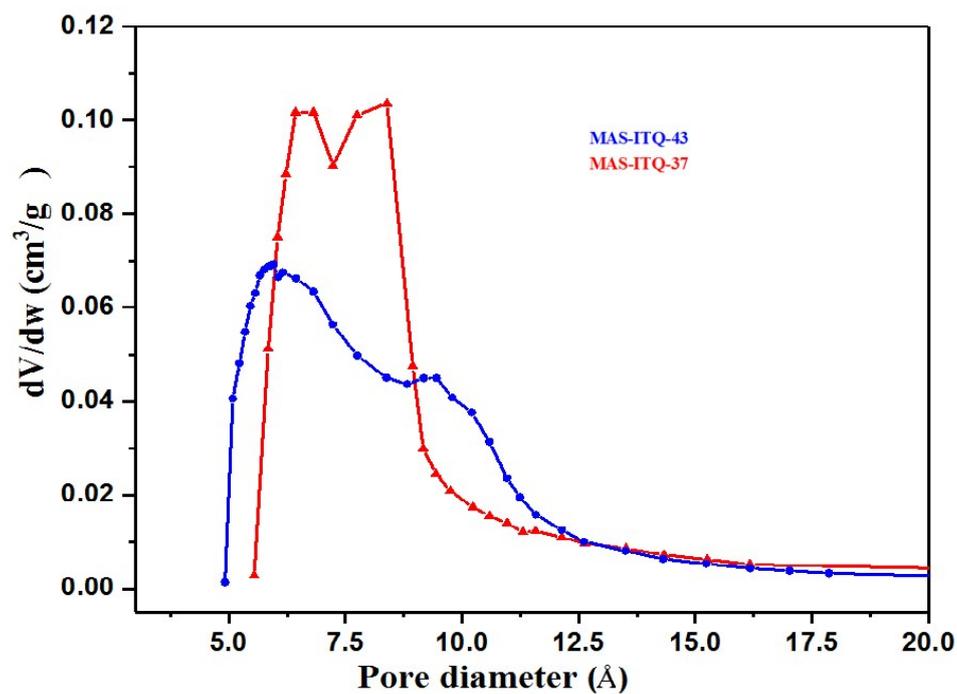
**Figure S3.**  $^{19}\text{F}$  MAS-NMR spectra of the as-synthesized MAS-ITQ-37 and MAS-ITQ-43.



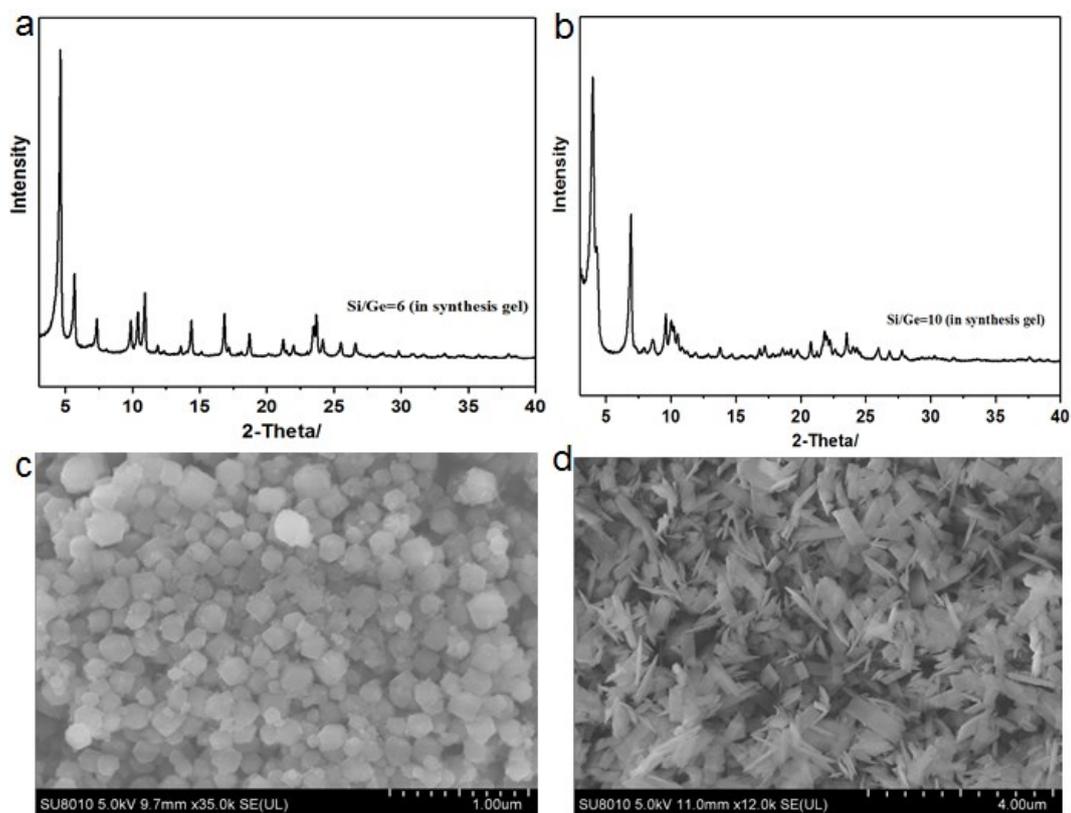
**Figure S4.**  $^{27}\text{Al}$  MAS-NMR spectrum of the as-synthesized MAS-ITQ-43.



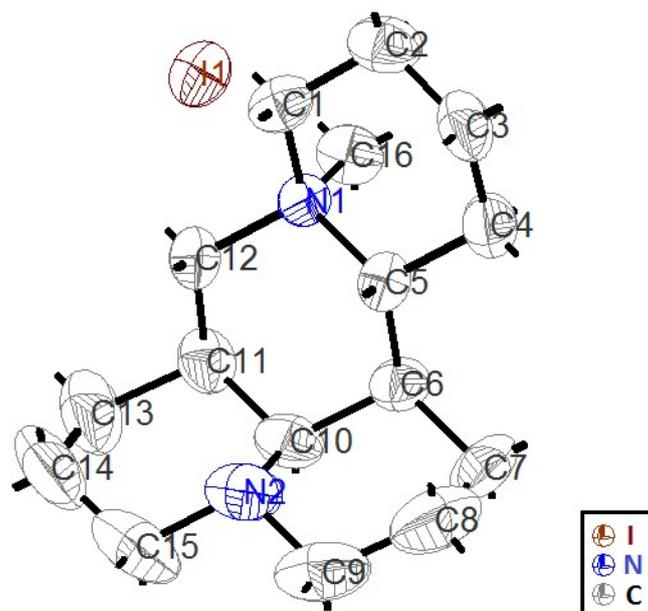
**Figure S5.** The TG curves of the as-synthesized MAS-ITQ-37 and MAS-ITQ-43.



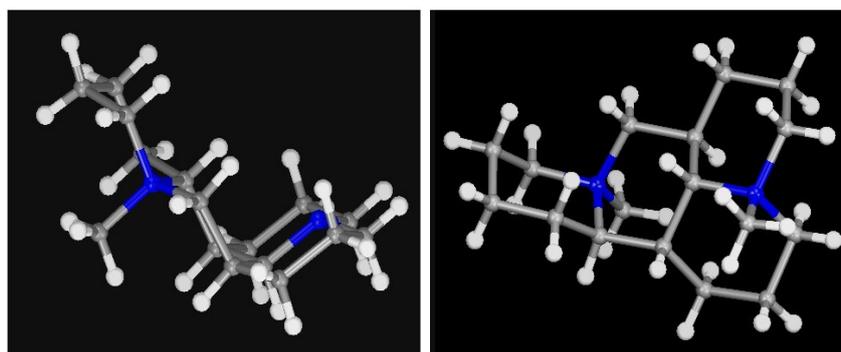
**Figure S6.** Experimental pore distributions of calcined MAS-ITQ-37 and MAS-ITQ-43 determined from N<sub>2</sub> adsorption.



**Figure S7.** PXRD patterns and SEM images of MAS-ITQ-37 (a, c) and MAS-ITQ-43 (b, d) prepared with the Si/Ge molar ratios of 6 and 10, respectively.



**Figure S8.** Representation of the asymmetric unit of MeMAS in its iodide showing ellipsoid at 50% probability, the hydrogen atoms are omitted for clarity.



**Figure S9.** Comparison of crystal structures of MeMAS (left) and Me<sub>2</sub>SOP (right).

**Table S1.** Chemical and elemental analyses of the as-made MAS-ITQ-37 and MAS-ITQ-43 synthesized in gels with Si/Ge= 2.

		MAS-ITQ-37	MAS-ITQ-43
<b>Chemical analyses (wt%)</b>	<b>Si</b>	<b>15.46</b>	<b>15.92</b>
	<b>Ge</b>	<b>28.4</b>	<b>23.6</b>
	<b>Al</b>	<b>none</b>	<b>undetectable</b>
<b>Elemental analyses (wt%)</b>	<b>C</b>	<b>16.99</b>	<b>21.12</b>
	<b>N</b>	<b>2.32</b>	<b>2.92</b>
	<b>H</b>	<b>3.33</b>	<b>3.3</b>
<b>Molar composition</b> <sup>a</sup>	$[(C_{16}H_{29}N_2)_{20}(H_2O)_{30}][Ge_{80}Si_{11}O_{400}H_{32}F_{20}]$	$[(C_{16}H_{29}N_2)_{3.7}(H_2O)_5][Ge_{11}Si_{18.7}O_{62.1}H_{5.4}F_{3.7}]$	

<sup>a</sup>The molar composition of the as-made MAS-ITQ-37 and MAS-ITQ-43 were derived by the combination of ICP, CHN elemental analyses, thermogravimetric analyses and crystallographic data of each zeolite structure.

**Table S2.** Crystal Data and Structure Refinement for compound MeMAS

Empirical formula	C <sub>16</sub> H <sub>29</sub> N <sub>2</sub> I
Formula weight	376.31
Temperature	293(2) K
Wavelength	0.71073 Å
Crystal system, space group	orthorhombic, <i>P</i> 2 <sub>1</sub> 2 <sub>1</sub> 2 <sub>1</sub>
Unit cell dimensions	$a = 9.045(3) \text{ \AA}$ $\alpha = 90^\circ$ $b = 18.898(6) \text{ \AA}$ $\beta = 90^\circ$ $c = 10.068(3) \text{ \AA}$ $\gamma = 90^\circ$
Volume	1721.1(10) Å <sup>3</sup>
<i>Z</i>	4
Absorption coefficient	1.853 mm <sup>-1</sup>
<i>F</i> (000)	768
Theta range for data collection	2.96 to 25.01°
Limiting indices	-10 ≤ <i>h</i> ≤ 10, -22 ≤ <i>k</i> ≤ 22, -11 ≤ <i>l</i> ≤ 11
Reflections collected / unique	3031 / 2202
<i>R</i> <sub>int</sub>	0.1695
Max. and min. transmission	0.8083 and 0.5552
Data / restraints / parameters	3031 / 0 / 173
Goodness-of-fit on <i>F</i> <sup>2</sup>	1.083
Final <i>R</i> indices [ <i>I</i> > 2σ( <i>I</i> )] <sup>a</sup>	<i>R</i> <sub>1</sub> = 0.0567, <i>wR</i> <sub>2</sub> = 0.1042
<i>R</i> indices (all data)	<i>R</i> <sub>1</sub> = 0.0952, <i>wR</i> <sub>2</sub> = 0.1202
Largest diff. peak and hole	1.011 and -0.662 e. Å <sup>-3</sup>

## Reference

[S1] M. E. C. P. Bruker AXS Inc., WI 53711-5373, USA 2000.

[S2] G. M. Sheldrick, *Acta Crystallogr. A* **2008**, *64*, 112-122.