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Redetermination at 113 K of 2,2-tetra-methylene-1,2-dihydroquinazolin-4(3H)-one

Lijun Zhang, Jiarong Li,* Daxin Shi and Jinnan Chen

 School of Chemical Engineering and the Environment, Beijing Institute of Technology, Beijing 100081, People's Republic of China
 Correspondence e-mail: jrli@bit.edu.cn

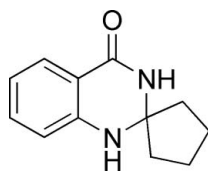
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 Key indicators: single-crystal X-ray study; $T = 113$ K; mean $\sigma(\text{C}-\text{C}) = 0.002$ Å; R factor = 0.041; wR factor = 0.104; data-to-parameter ratio = 16.9.

The title compound {systematic name: spiro[cyclopentane-1,2'(1*H*)-quinazolin]-4'(3'*H*)-one]}, $\text{C}_{12}\text{H}_{14}\text{N}_2\text{O}$, has been reported previously [Klemm, Weakley, Gilbertson & Song (1998). *J. Heterocycl. Chem.* **35**, 1269–1273]. Its structure has been redetermined at 113 K with greater precision for all data. The molecule is built up from two fused six-membered rings and one five-membered ring linked through a spiro C atom. The pyrimidine ring has an envelope conformation and the cyclopentane ring adopts a distorted boat form. There are intermolecular $\text{N}-\text{H}\cdots\text{O}$ hydrogen bonds, which form a two-dimensional sheet parallel to the (001) plane.

Related literature

For related literature, see: Bernstein *et al.* (1995); Cremer & Pople (1975); Etter *et al.* (1990); Shi *et al.* (2004); Summers *et al.* (1986).



Experimental

Crystal data

$\text{C}_{12}\text{H}_{14}\text{N}_2\text{O}$
 $M_r = 202.25$
 Orthorhombic, *Pbca*
 $a = 10.3872$ (12) Å
 $b = 12.0252$ (13) Å
 $c = 16.3027$ (19) Å

$V = 2036.3$ (4) Å³
 $Z = 8$
 Mo $K\alpha$ radiation
 $\mu = 0.09$ mm⁻¹
 $T = 113$ (2) K
 $0.26 \times 0.24 \times 0.16$ mm

Data collection

Rigaku Saturn diffractometer
 Absorption correction: multi-scan
 (Jacobson, 1998)
 $T_{\min} = 0.972$, $T_{\max} = 0.984$

23533 measured reflections
 2403 independent reflections
 2200 reflections with $I > 2\sigma(I)$
 $R_{\text{int}} = 0.033$

Refinement

$R[F^2 > 2\sigma(F^2)] = 0.041$
 $wR(F^2) = 0.104$
 $S = 1.09$
 2403 reflections
 142 parameters
 2 restraints

H atoms treated by a mixture of independent and constrained refinement
 $\Delta\rho_{\text{max}} = 0.31$ e Å⁻³
 $\Delta\rho_{\text{min}} = -0.23$ e Å⁻³

Table 1

Hydrogen-bond geometry (Å, °).

$D-\text{H}\cdots A$	$D-\text{H}$	$\text{H}\cdots A$	$D\cdots A$	$D-\text{H}\cdots A$
$\text{N1}-\text{H1}\cdots\text{O1}^{\text{i}}$	0.857 (9)	2.043 (9)	2.8936 (12)	171.2 (14)
$\text{N2}-\text{H2}\cdots\text{O1}^{\text{ii}}$	0.857 (9)	2.077 (9)	2.9303 (13)	173.3 (14)

Symmetry codes: (i) $-x, -y + 1, -z + 1$; (ii) $-x + \frac{1}{2}, y - \frac{1}{2}, z$.

Data collection: *CrystalClear* (Rigaku, 2004); cell refinement: *CrystalClear*; data reduction: *CrystalClear*; program(s) used to solve structure: *SHELXS97* (Sheldrick, 1997); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics: *ORTEP-III* (Burnett & Johnson, 1996), *ORTEP-3 for Windows* (Farrugia, 1997) and *CAMERON* (Watkin *et al.*, 1993); software used to prepare material for publication: *SHELXL97*.

We thank Beijing Institute of Technology for financial support and Naikai University for the X-ray diffraction analysis.

Supplementary data and figures for this paper are available from the IUCr electronic archives (Reference: DN2298).

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supporting information

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Redetermination at 113 K of 2,2-tetramethylene-1,2-dihydroquinazolin-4(3H)-one

Lijun Zhang, Jiarong Li, Daxin Shi and Jinnan Chen

S1. Comment

When we used 2-aminobenzonitrile and cyclopentanone in presence of zinc chloride to synthesize tacrine (Summers *et al.*, 1986) derivative, the unexpected spiro compound (I) (Scheme 1), was obtained. Its structure has been already reported (Klemm *et al.*, 1998), however, as we were investigating the molecular and supramolecular architecture of related compounds, its structural redetermination at lower temperature (113 K) has been undertaken

The molecular structure of (I) is built up with two fused six membered ring and a five membered ring linked through a spiro C atom (Fig. 1). The pyrimidine ring has an envelope conformation with puckering parameters $Q=0.3821(11)$ Å, $\Theta=115.21(16)^\circ$ and $\varphi=108.70(19)^\circ$ (Cremer & Pople, 1975). The five-membered ring displays an envelope conformation at C12 with puckering parameters $Q(2)=0.3925(15)$ Å and $\varphi(2)=319.8(2)\%$. The geometry of the fused rings compares well with the related 3-phenyl-1,2-dihydroquinazolin-4(3H)-one derivative (Shi *et al.*, 2004).

The crystal structure of (I) is stabilized by the interplay of N—H \cdots O interactions. The two N—H groups form N—H \cdots O hydrogen bonds with the ketone O atom of symmetry related molecules building a $R_2^2(8)$ graph set motif (Etter *et al.*, 1990; Bernstein *et al.*, 1995). Those motifs formed with N—H \cdots O hydrogen bonds link to each other building a two dimensionnal network parallel to the (0 0 1) plane (Fig. 2, Table 1).

S2. Experimental

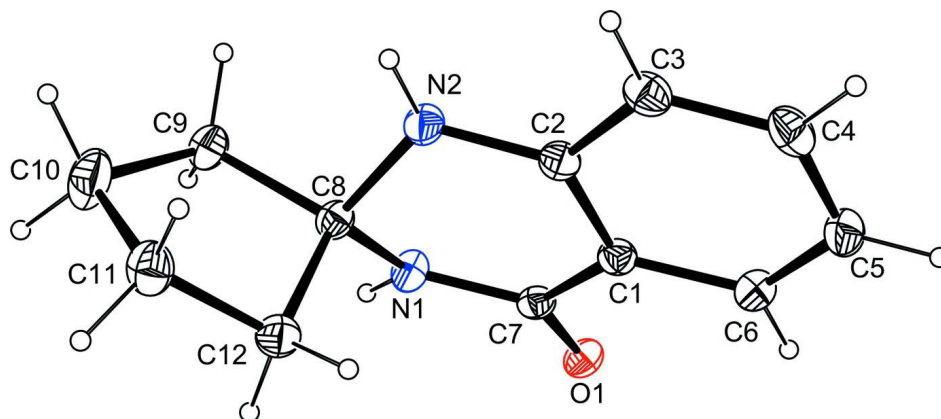
2,2-Tetramethylene-1,2-dihydroquinazolin-4(3H)-one (I) was prepared from the reflux of 2-aminobenzonitrile (1 mmol) with cyclopentanone (1 mmol) in presence of zinc chloride (1.2 mmol) in 10 ml DMF for 1.5 h. Then the reaction mixture was cooled and quenched with water and the precipitate was separated by filtration. The filtration residue was dispersed into water and titrated to pH 12–13 by 20% sodium hydroxide. After filtration, the product was obtained in 70% yield by column chromatography (200–300 mesh silica gel, ethyl acetate–petroleum with 1:2).

The single-crystal of (I) was cultured from a solution of ethanol by slow evaporation at room temperature.

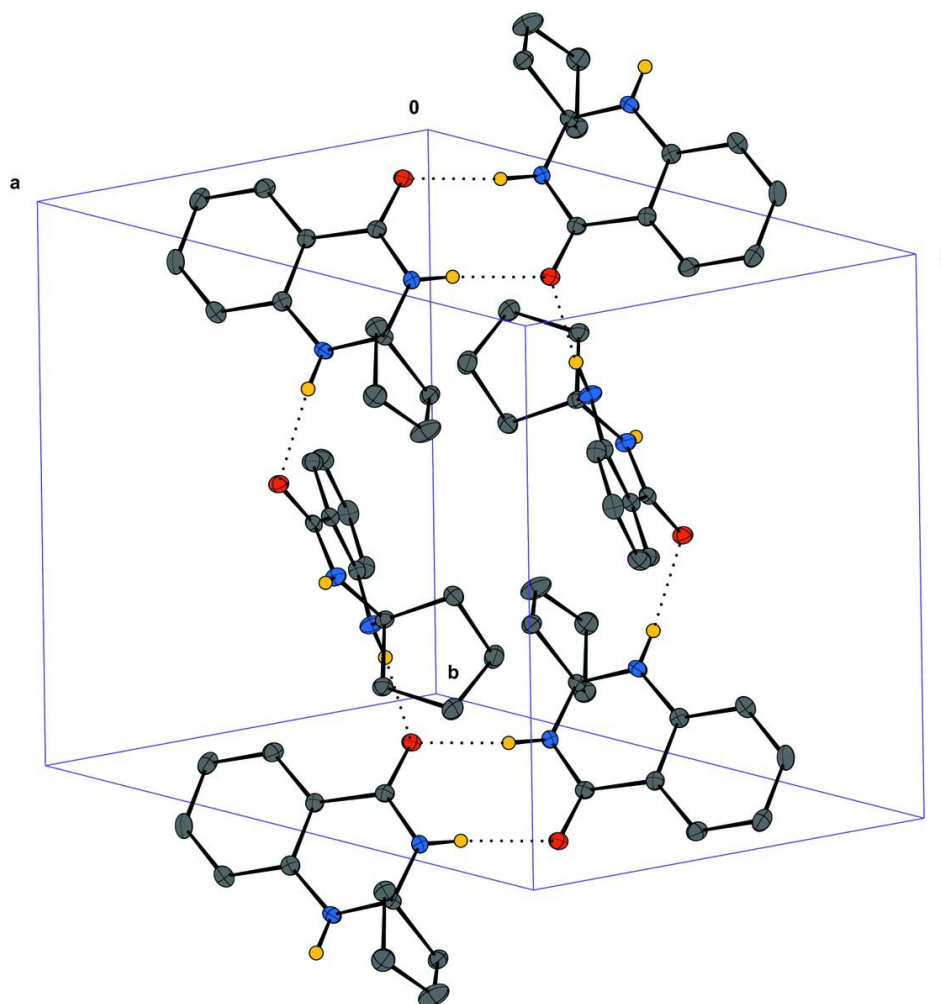
Spectra data: IR (KBr, cm^{-1}): 3289, 3166, 2934, 1638, 1613, 1429; ^1H NMR (DMSO- d_6) δ_{H} : 1.75–2.08 (8H, m, C_4H_8), 6.07 (1H, s, NH), 6.73 (2H, dd, $J=7.8, 8.0$ Hz, ArH), 7.19 (1H, s, NH), 7.24–7.26 (1H, m, $J=7.2$ Hz, ArH), 7.73 (1H, d, $J=8.0$ Hz, ArH); ^{13}C NMR (DMSO- d_6) δ_{C} : 21.97 (2 C), 38.88 (2 C), 77.05, 114.32, 114.57, 116.53, 127.23, 132.99, 147.49, 163.42; MS (ESI): m/z (%) = 203.1 (100) [$M+H$] $^+$; $\text{C}_{12}\text{H}_{14}\text{N}_2\text{O}$: calcd. C 71.26, H 6.98, N 13.85; found C 71.38, H 6.71, N 13.49.

S3. Refinement

All H atoms attached to C atoms were fixed geometrically and treated as riding with C—H = 0.93 Å (aromatic) or 0.97 Å (methylene) with $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{C or N})$. H atoms of NH group were located in difference Fourier maps and included in the subsequent refinement using restraints (N—H = 0.85 (1) Å) with $U_{\text{iso}}(\text{H}) = 1.2U_{\text{eq}}(\text{N})$.

**Figure 1**

The molecular structure of (I), with the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level. H atoms are represented as small spheres of arbitrary radii.

**Figure 2**

Partial packing showing one sheet of molecules connected by N—H...O hydrogen bonds (dashed lines). H atoms not involved in hydrogen bonding have been omitted for clarity.

spiro[cyclopentane-1,2'(1'H)-quinazolin]-4'(3'H)-one]

Crystal data

C₁₂H₁₄N₂O $M_r = 202.25$ Orthorhombic, *Pbca*

Hall symbol: -P 2ac 2ab

 $a = 10.3872$ (12) Å $b = 12.0252$ (13) Å $c = 16.3027$ (19) Å $V = 2036.3$ (4) Å³ $Z = 8$ $F(000) = 864$ $D_x = 1.319$ Mg m⁻³Mo $K\alpha$ radiation, $\lambda = 0.71070$ Å

Cell parameters from 5598 reflections

 $\theta = 2.6$ – 27.9° $\mu = 0.09$ mm⁻¹ $T = 113$ K

Prism, colorless

 $0.26 \times 0.24 \times 0.16$ mm

Data collection

Rigaku Saturn

diffractometer

Radiation source: rotating anode

Confocal monochromator

Detector resolution: 7.31 pixels mm⁻¹ ω scans

Absorption correction: multi-scan

(Jacobson, 1998)

 $T_{\min} = 0.972$, $T_{\max} = 0.984$

23533 measured reflections

2403 independent reflections

2200 reflections with $I > 2\sigma(I)$ $R_{\text{int}} = 0.033$ $\theta_{\max} = 27.9^\circ$, $\theta_{\min} = 2.9^\circ$ $h = -13 \rightarrow 13$ $k = -15 \rightarrow 15$ $l = -21 \rightarrow 21$

Refinement

Refinement on F^2

Least-squares matrix: full

 $R[F^2 > 2\sigma(F^2)] = 0.041$ $wR(F^2) = 0.104$ $S = 1.09$

2403 reflections

142 parameters

2 restraints

Primary atom site location: structure-invariant

direct methods

Secondary atom site location: difference Fourier map

Hydrogen site location: inferred from neighbouring sites

H atoms treated by a mixture of independent and constrained refinement

 $w = 1/[\sigma^2(F_o^2) + (0.0476P)^2 + 0.9123P]$ where $P = (F_o^2 + 2F_c^2)/3$ $(\Delta/\sigma)_{\max} = 0.001$ $\Delta\rho_{\max} = 0.31$ e Å⁻³ $\Delta\rho_{\min} = -0.23$ e Å⁻³

Special details

Geometry. All e.s.d.'s (except the e.s.d. in the dihedral angle between two l.s. planes) are estimated using the full covariance matrix. The cell e.s.d.'s are taken into account individually in the estimation of e.s.d.'s in distances, angles and torsion angles; correlations between e.s.d.'s in cell parameters are only used when they are defined by crystal symmetry. An approximate (isotropic) treatment of cell e.s.d.'s is used for estimating e.s.d.'s involving l.s. planes.

Refinement. Refinement of F^2 against ALL reflections. The weighted R -factor wR and goodness of fit S are based on F^2 , conventional R -factors R are based on F , with F set to zero for negative F^2 . The threshold expression of $F^2 > \sigma(F^2)$ is used only for calculating R -factors(gt) etc. and is not relevant to the choice of reflections for refinement. R -factors based on F^2 are statistically about twice as large as those based on F , and R -factors based on ALL data will be even larger.

Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å²)

	x	y	z	$U_{\text{iso}}^*/U_{\text{eq}}$
O1	0.08554 (8)	0.58043 (6)	0.58072 (5)	0.01704 (19)
N1	0.14762 (9)	0.42225 (8)	0.51639 (6)	0.0162 (2)
H1	0.0821 (11)	0.4272 (12)	0.4846 (8)	0.019*

N2	0.30566 (10)	0.30494 (8)	0.57191 (6)	0.0176 (2)
H2	0.3437 (13)	0.2417 (9)	0.5745 (9)	0.021*
C1	0.27524 (10)	0.48724 (9)	0.62879 (6)	0.0149 (2)
C2	0.34067 (10)	0.38442 (9)	0.62795 (7)	0.0155 (2)
C3	0.43548 (11)	0.36400 (10)	0.68791 (7)	0.0189 (2)
H3	0.4778	0.2959	0.6891	0.023*
C4	0.46564 (11)	0.44475 (10)	0.74485 (7)	0.0208 (3)
H4	0.5295	0.4308	0.7835	0.025*
C5	0.40171 (12)	0.54737 (10)	0.74554 (7)	0.0210 (3)
H5	0.4232	0.6013	0.7840	0.025*
C6	0.30602 (11)	0.56750 (9)	0.68820 (7)	0.0186 (2)
H6	0.2618	0.6347	0.6891	0.022*
C7	0.16430 (10)	0.50191 (9)	0.57324 (7)	0.0144 (2)
C8	0.24515 (11)	0.33881 (9)	0.49553 (6)	0.0148 (2)
C9	0.18435 (11)	0.23692 (10)	0.45265 (7)	0.0182 (2)
H9A	0.1703	0.1774	0.4918	0.022*
H9B	0.1023	0.2568	0.4283	0.022*
C10	0.27944 (13)	0.20002 (11)	0.38614 (9)	0.0300 (3)
H10A	0.3038	0.1229	0.3942	0.036*
H10B	0.2410	0.2075	0.3322	0.036*
C11	0.39717 (12)	0.27571 (10)	0.39391 (8)	0.0229 (3)
H11A	0.4620	0.2424	0.4290	0.027*
H11B	0.4348	0.2906	0.3406	0.027*
C12	0.34353 (11)	0.38216 (10)	0.43211 (7)	0.0195 (2)
H12A	0.3019	0.4282	0.3911	0.023*
H12B	0.4111	0.4248	0.4585	0.023*

Atomic displacement parameters (Å²)

	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0156 (4)	0.0139 (4)	0.0217 (4)	0.0012 (3)	-0.0010 (3)	-0.0011 (3)
N1	0.0137 (4)	0.0154 (4)	0.0194 (5)	0.0023 (3)	-0.0038 (4)	-0.0026 (4)
N2	0.0213 (5)	0.0138 (5)	0.0178 (5)	0.0050 (4)	-0.0044 (4)	-0.0006 (4)
C1	0.0145 (5)	0.0152 (5)	0.0149 (5)	-0.0015 (4)	0.0004 (4)	0.0010 (4)
C2	0.0144 (5)	0.0165 (5)	0.0155 (5)	-0.0010 (4)	0.0016 (4)	0.0010 (4)
C3	0.0180 (5)	0.0210 (5)	0.0178 (5)	0.0034 (4)	0.0003 (4)	0.0027 (4)
C4	0.0179 (5)	0.0287 (6)	0.0157 (5)	-0.0010 (5)	-0.0031 (4)	0.0020 (5)
C5	0.0231 (6)	0.0220 (6)	0.0180 (5)	-0.0051 (5)	-0.0032 (4)	-0.0020 (4)
C6	0.0204 (6)	0.0161 (5)	0.0194 (6)	-0.0017 (4)	-0.0002 (4)	-0.0007 (4)
C7	0.0140 (5)	0.0132 (5)	0.0159 (5)	-0.0015 (4)	0.0018 (4)	0.0015 (4)
C8	0.0141 (5)	0.0134 (5)	0.0168 (5)	0.0015 (4)	-0.0010 (4)	-0.0008 (4)
C9	0.0164 (5)	0.0171 (5)	0.0212 (5)	-0.0009 (4)	-0.0007 (4)	-0.0035 (4)
C10	0.0310 (7)	0.0266 (7)	0.0324 (7)	-0.0065 (5)	0.0113 (6)	-0.0128 (5)
C11	0.0196 (6)	0.0256 (6)	0.0235 (6)	0.0013 (5)	0.0037 (5)	-0.0038 (5)
C12	0.0199 (6)	0.0187 (6)	0.0198 (5)	-0.0028 (4)	0.0021 (4)	0.0002 (4)

Geometric parameters (Å, °)

O1—C7	1.2553 (13)	C5—H5	0.9300
N1—C7	1.3442 (14)	C6—H6	0.9300
N1—C8	1.4659 (14)	C8—C12	1.5443 (15)
N1—H1	0.857 (9)	C8—C9	1.5456 (15)
N2—C2	1.3712 (15)	C9—C10	1.5323 (17)
N2—C8	1.4531 (14)	C9—H9A	0.9700
N2—H2	0.857 (9)	C9—H9B	0.9700
C1—C6	1.4041 (15)	C10—C11	1.5297 (17)
C1—C2	1.4110 (15)	C10—H10A	0.9700
C1—C7	1.4762 (15)	C10—H10B	0.9700
C2—C3	1.4091 (16)	C11—C12	1.5287 (17)
C3—C4	1.3794 (16)	C11—H11A	0.9700
C3—H3	0.9300	C11—H11B	0.9700
C4—C5	1.4014 (17)	C12—H12A	0.9700
C4—H4	0.9300	C12—H12B	0.9700
C5—C6	1.3857 (16)		
C7—N1—C8	123.96 (9)	N1—C8—C12	112.43 (9)
C7—N1—H1	118.0 (9)	N2—C8—C9	110.01 (9)
C8—N1—H1	117.1 (10)	N1—C8—C9	111.42 (9)
C2—N2—C8	119.36 (9)	C12—C8—C9	103.60 (9)
C2—N2—H2	117.6 (10)	C10—C9—C8	106.63 (9)
C8—N2—H2	119.3 (10)	C10—C9—H9A	110.4
C6—C1—C2	119.95 (10)	C8—C9—H9A	110.4
C6—C1—C7	121.25 (10)	C10—C9—H9B	110.4
C2—C1—C7	118.35 (10)	C8—C9—H9B	110.4
N2—C2—C3	121.74 (10)	H9A—C9—H9B	108.6
N2—C2—C1	119.31 (10)	C11—C10—C9	106.53 (10)
C3—C2—C1	118.85 (10)	C11—C10—H10A	110.4
C4—C3—C2	120.19 (11)	C9—C10—H10A	110.4
C4—C3—H3	119.9	C11—C10—H10B	110.4
C2—C3—H3	119.9	C9—C10—H10B	110.4
C3—C4—C5	121.17 (11)	H10A—C10—H10B	108.6
C3—C4—H4	119.4	C12—C11—C10	103.92 (10)
C5—C4—H4	119.4	C12—C11—H11A	111.0
C6—C5—C4	119.23 (11)	C10—C11—H11A	111.0
C6—C5—H5	120.4	C12—C11—H11B	111.0
C4—C5—H5	120.4	C10—C11—H11B	111.0
C5—C6—C1	120.57 (11)	H11A—C11—H11B	109.0
C5—C6—H6	119.7	C11—C12—C8	103.37 (9)
C1—C6—H6	119.7	C11—C12—H12A	111.1
O1—C7—N1	121.26 (10)	C8—C12—H12A	111.1
O1—C7—C1	122.62 (10)	C11—C12—H12B	111.1
N1—C7—C1	116.01 (9)	C8—C12—H12B	111.1
N2—C8—N1	106.98 (9)	H12A—C12—H12B	109.1
N2—C8—C12	112.46 (9)		

Hydrogen-bond geometry (Å, °)

<i>D</i> —H \cdots <i>A</i>	<i>D</i> —H	H \cdots <i>A</i>	<i>D</i> \cdots <i>A</i>	<i>D</i> —H \cdots <i>A</i>
N1—H1 \cdots O1 ⁱ	0.86 (1)	2.04 (1)	2.8936 (12)	171 (1)
N2—H2 \cdots O1 ⁱⁱ	0.86 (1)	2.08 (1)	2.9303 (13)	173 (1)

Symmetry codes: (i) $-x, -y+1, -z+1$; (ii) $-x+1/2, y-1/2, z$.