Crystal Structure of 2,3,7-Tris(cyclohexylimino)-3,4-dihydro-4-spirocycloheptan-2*H*-furo[3,4-*b*]pyran-5(7*H*)-one

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The title compound crystallized in triclinic space group $P\bar{1}$ with cell parameters $a = 6.6634(3)\text{\AA}$, $b = 12.3296(6)\text{\AA}$, $c = 17.0338(8)\text{\AA}$, $\alpha = 86.394(1)^\circ$, $\beta = 81.052(1)^\circ$, $\gamma = 85.002(1)^\circ$, $V = 1375.43(11)\text{\AA}^3$, $Z = 2$, $D_{\text{cal}} = 1.226\text{ Mg/m}^3$ at $T = 100(2)\text{K}$. The structure was refined by full-matrix least-squares procedures to find $R_1 = 0.0480$ and $wR_2 = 0.1143$ for 5663 reflections. All atoms in the furo-pyran ring are in a plane, except C_6 . All three cyclohexyl rings of isocyanides adapt chair conformation, and their plane alignment is noncoplanar with respect to furo-pyran plane. The spiro cycloheptyl ring has a chair conformation.

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Multi-component reactions (MCRs) have been extensively investigated in organic and diversity oriented synthesis, primarily due to their ability to generate complex functionality from simple starting materials in a one-step transformations.^{1,2} Great efforts have been, and still are made to find and develop new multi-component reactions. An isocyanide-based multi-component reaction has a magic situation for providing a diversity oriented synthesis. Here, we introduce the crystal structure product of a multi-component (3+1) reaction of isocyanide and alkylidene Meldrum's acid.³⁻⁶

The title compound was prepared by the reaction of cyclohexyl isocyanide and alkylidene Meldrum's acid (3:1 ratio) in dichloromethane at room temperature within four hours (Fig. 1). After evaporating the solvent, ethanol was added to the residue, and a white powder was separated from the reaction mixture. More purification and recrystallization was done in hot ethanol.

The crystal structure (Fig. 2) was solved by direct methods and refined on F^2 by full-matrix least-squares procedures. The title compound crystallized in triclinic space group $P\bar{1}$ with cell parameters $a = 6.6634(3)\text{\AA}$, $b = 12.3296(6)\text{\AA}$, $c = 17.0338(8)\text{\AA}$, $\alpha = 86.394(1)^\circ$, $\beta = 81.052(1)^\circ$, $\gamma = 85.002(1)^\circ$, $V = 1375.43(11)\text{\AA}^3$, $Z = 2$, $D_{\text{cal}} = 1.226\text{ Mg/m}^3$ at $T = 100(2)\text{K}$. The non-hydrogens were refined anisotropically, and all of the hydrogens were fixed as a riding model over their heavier atoms.

Fig. 1 Chemical diagram of the title compound.

A fine cycle refinement [$I > 2\sigma(I)$] converged to $R_1 = 0.0480$ and $wR_2 = 0.1143$ for the observed reflections [$I > 2\sigma(I)$] is 5663. Crystal data and other relevant parameters for the compound are given in Table 1. All atoms in the furo-pyran ring are in a plane; except for C_6 , all three cyclohexyl rings of isocyanides adapt to a chair conformation, and their plane aligns in a noncoplanar way with respect to the furo-pyran plane. The spiro cycloheptyl ring has a chair conformation. The bond lengths of the furo-pyran atoms are normal. Although the imine bond lengths of $N(1)-C(2)$ and $N(2)-C(5)$ are similar, the bond lengths of $N(3)-C(6)$ show a slight elongation. As a result, these bond lengths

Table 1 Crystal and experimental data

Chemical formula: $C_{31}H_{45}N_3O_3$	
Formula weight = 507.70	
$T = 100(2)\text{K}$	
Crystal system: triclinic	Space group: $P\bar{1}$
$a = 6.6634(3)\text{\AA}$	$\alpha = 86.394(1)^\circ$
$b = 12.3296(6)\text{\AA}$	$\beta = 81.052(1)^\circ$
$c = 17.0338(8)\text{\AA}$	$\gamma = 85.002(1)^\circ$
$V = 1375.43(11)\text{\AA}^3$	$Z = 2$
$D_x = 1.226\text{ Mg/m}^3$	
Radiation: Mo K_α ($\lambda = 0.71073\text{\AA}$)	
$\mu(\text{Mo } K_\alpha) = 0.079\text{ mm}^{-1}$	$F(0\ 0\ 0) = 552$
Crystal size = $0.44 \times 0.12 \times 0.10\text{ mm}^3$	
No. of reflections collected = 16875	
No. of independent reflections = 7286 [$R(\text{int}) = 0.0287$]	
θ range for data collection: 1.21 to 29.00°	
Data/Restraints/Parameters = 7286/0/334	
Goodness-of-fit on $F^2 = 1.023$	
R indices [$I > 2\sigma(I)$]: $R_1 = 0.0480$, $wR_2 = 0.1143$	
R indices (all data): $R_1 = 0.0656$, $wR_2 = 0.1252$	
$\Delta\rho = 0.001$	
$(\Delta/\sigma)_{\text{max}} = 0.59\text{ e/\AA}^{-3}$	
$(\Delta/\sigma)_{\text{min}} = -0.58\text{ e/\AA}^{-3}$	
Measurment: Bruker APEX-II CCD	
Program System: SHELXL-97, SADABS	
Structure Determination: SHELXS-97	
CCDC deposition number: 686152	

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Table 2 Selected bond lengths [Å] and angles [°] for the compound

O1-C2	1.3975(15)	O1-C1-C4	108.34(10)
O1-C1	1.4004(15)	N1-C2-O1	126.32(11)
O2-C1	1.1953(16)	N1-C2-C3	127.88(12)
O3-C3	1.3503(15)	O1-C2-C3	105.72(10)
O3-C5	1.4206(14)	C4-C3-O3	127.26(11)
N1-C2	1.2551(17)	C4-C3-C2	110.99(11)
N1-C14	1.4731(15)	O3-C3-C2	121.72(11)
N2-C5	1.2493(16)	C3-C4-C1	105.99(11)
N3-C6	1.2676(16)	C3-C4-C7	123.72(11)
N3-C26	1.4667(17)	O3-C5-C6	115.06(10)
C1-C4	1.4743(17)	N3-C6-C5	124.09(11)
C2-C3	1.4695(17)	N3-C6-C7	122.03(11)
C3-C4	1.3375(17)	C5-C6-C7	113.88(10)
C4-C7	1.5139(17)	C4-C7-C6	101.65(10)
C5-C6	1.5124(17)	C4-C7-C8	110.70(10)
C6-C7	1.5397(17)	C6-C7-C8	111.75(10)
		C8-C7-C13	115.20(11)
		C9-C8-C7	119.14(12)
		N1-C14-C19	110.27(10)
		N1-C14-C15	106.97(10)
		C19-C14-C15	110.36(10)
		C16-C15-C14	111.25(11)
		C17-C16-C15	111.10(11)
		C18-C17-C16	111.46(11)
		C7-C18-C19	110.96(11)

are similar to those in other reports previously published,⁷ in which for the imine bond there is no resonance effect, or a very weak resonance effect may be exist. The plane of the three cyclohexyl rings aligns in a noncoplanar way with respect to the furo-pyran plane, due to a reduced repulsion of the crowded structure. The dihedral torsion angle of N(2)-C(5)-C(6)-N(3) is 54.29°.

References

1. I. Ugi, *Angew. Chem. Int. Ed. Engl.*, **1982**, 21, 810.

Fig. 2 Molecular structure (ORTEP-III plot) along with an arbitrary numbering of the atoms (thermal ellipsoids for non-hydrogen atoms).

2. A. Domling and I. Ugi, *Angew. Chem. Int. Ed. Engl.*, **2000**, 39, 3168.
3. A. N. Meldrum, *J. Chem. Soc.*, **1908**, 93, 598.
4. D. Davidson and S. A. Bernhard, *J. Am. Chem. Soc.*, **1948**, 70, 3426.
5. B. C. Chen, *Heterocycles*, **1991**, 32, 529. H. McNab, *Chem. Soc. Rev.*, **1978**, 7, 354.
6. I. Yavari and A. Habibi, *Synthesis*, **2004**, 7, 989 and references cited therein.
7. H. Hartung, A. Stettler, and W. Weissflog, *J. Mole. Struct.*, **2000**, 526, 31; I. Kilic, F. Ersahin, E. Agar, and S. Isik, *X-Ray Struct. Anal. Online*, **2009**, 25, 25; J. Li, Z.-P. Liang, F.-T. Kong, H. Zhang, and X.-S. Tai, *Chinese J. Struct. Chem.*, **2009**, 28(5), 577.