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Benz[4,5]isoquino[1,2-*b*]quinazoline-7,9-dione and a Rearrangement Product of its Hydrolysis, 2-(1,8-Naphthalenedicarboximido)benzamide

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Abstract

Benz[4,5]isoquino[1,2-*b*]quinazoline-7,9-dione, (1), $C_{19}H_{10}N_2O_2$, was isolated as a product of the reaction between naphthalic anhydride and anthranilamide after crystallization from dimethylformamide. Recrystallization from concentrated (95%) formic acid resulted in 2-(1,8-naphthalenedicarboximido)benzamide, (2), $C_{19}H_{12}N_2O_3$, as a rearrangement product of hydrolysis [alternative systematic name: 2-(1,3-dioxo-2,3-dihydro-1*H*-benz[*de*]isoquinol-2-yl)benzamide]. The two crystallographically independent molecules of (1) [(1*a*) and (1*b*)] are substantially non-planar as a result of strong intramolecular steric repulsion between their *cis*-arranged carbonyl groups [the O...O distances are 2.584 (3) and 2.664 (3) Å, and the dihedral angles between the naphthalene and benzene nuclei are 8.75 (7) and 14.46 (7)°, respectively]. Molecule (2) is also sterically hindered with an approximately orthogonal orientation of the naphthalenedicarboximide and *o*-phenylene groups [dihedral angle 87.05 (4)°] and absence of π -conjugation between the *o*-phenylene and amide groups [dihedral angle 40.02 (6)°].

Comment

As part of our investigations into the synthesis of regular polynaphthoquinazolones from bis-naphthalic anhydrides and bis-anthranylamides, the model reaction between naphthalic anhydride and anthranilamide has been carried out (see scheme below) (Ponomarev, Skuratova, Lindeman, Sinichkin, Vinogradova & Rusanov, 1994).

Only one of the two possible isomeric products, benz-[4,5]isoquino[1,2-*b*]quinazoline-7,9-dione, (1), was isolated from the reaction mixture by crystallization from dimethylformamide. Recrystallization from concentrated (95%) formic acid resulted in the formation of 2-(1,8-naphthalenedicarboximido)benzamide, (2), which may be considered as a rearrangement product of the hydrolysis of (1).

The two crystallographically independent molecules of (1) [(1*a*) and (1*b*) in Fig. 1] are substantially distorted relative to the ideal π -conjugated planar structure. The dihedral angles between the planes of the naphthalene and benzene nuclei are 8.75 (7)° for (1*a*) and 14.46 (7)° for (1*b*). This results from bending of the non-aromatic rings: the diazoline ring of the quinazoline moiety adopts a sofa conformation [atoms N1 and N1*a* are shifted from the planes of the related benzene rings by 0.106 and 0.208 (4) Å, respectively, whereas the deviation of the other atoms does not exceed 0.044 (3) Å]. The tetrahydropyridine ring of the isoquinoline moiety has a boat-like conformation with atoms N1 (N1*a*) and C12 (C12*a*) shifted by 0.194 (3) Å [0.258 (3) Å] and 0.103 (3) Å [0.104 (3) Å] from the mean plane of the naphthalene moiety; atom C11 (C11*a*) deviates by 0.085 (3) Å [0.034 (3) Å] in the opposite direction.

These distortions are caused principally by forced short intramolecular contacts between *cis*-disposed carbonyl groups: the O...O distances are 2.584 and 2.664 (3) Å in molecules (1*a*) and (1*b*), respectively (at such a mutual orientation of the polar carbonyl groups, an additional electrostatic repulsion of their dipoles evidently takes place). In addition, the short intramolecular contacts O1...H—C7 [2.57 (3) and 2.55 (3) Å in molecules (1*a*) and (1*b*), respectively], O1'...H—C6' [2.56 (3) and 2.60 (3) Å] and N2...H—C2 [2.40 (3) and 2.46 (3) Å], are also seemingly of the repulsive type. As a result, atoms O1 and N2 deviate in opposite directions from the mean plane of the naphthalene moiety

by 0.404 (3) and −0.117 (3) Å in (1*a*) and by 0.293 (3) and −0.088 (3) Å in (1*b*), and atom O1' deviates from the mean plane of the phenylene moiety by 0.186 (4) in (1*a*) and 0.085 (4) Å in (1*b*).

Fig. 1. Perspective view of the two crystallographically independent molecules of (1) [top (1*a*), bottom (1*b*) in comparable orientations [molecule (1*b*) is shown inverted relative to the table of coordinates] showing the labelling of the non-H atoms. Displacement ellipsoids are shown at the 50% probability level.

The noticeable difference in the degree of non-planarity for the two crystallographically independent molecules (1*a*) and (1*b*) seems to result from their different crystal environments. In (1), the molecules are packed in stacks [$\cdots(1b')\cdots(1b)\cdots(1a')\cdots(1a)\cdots$]_{*n*} along [101] (Fig. 2). In the stacks, (1*a*) is overlapped fully and closely (interplanar distances are *ca* 3.42 Å)

Fig. 2. Perspective view of a stack of molecules of (1) in the crystal.

by two antiparallel neighbouring molecules ($1b''$) and ($1a'$), but only the naphthalenedicarboximide moiety is overlapped by the packing of the antiparallel ($1b$) and ($1b'$) molecules, where the interplanar distance is *ca* 3.53 Å.

Molecule (2), to our knowledge, is the first aryl-substituted 1,8-naphthalenedicarboximide to be investigated structurally. As a result of steric restrictions enforced by the two carbonyl groups, the benzamide moiety is almost perpendicular to the planar naphthalenedicarboximide moiety [dihedral angle 87.05 (4)°; Fig. 3]. The N1—C2' bond length of 1.449 (1) Å is longer than the standard value of 1.371 Å (Allen, Kennard, Watson, Brammer, Orpen & Taylor, 1987). The amide group is also twisted around the C1'—C7' bond [by 40.02 (6)°] such that it is not π -conjugated with the benzene ring. Nevertheless, the length of this bond [1.502 (2) Å] is normal (Allen *et al.*, 1987).

Fig. 3. Perspective view of molecule (2) showing the labelling of the non-H atoms. Displacement ellipsoids are shown at the 50% probability level.

The bond-length distribution in the naphthalenedicarboximide moieties of (1) and (2) is very similar to that found earlier in *N*-ethyl-1,8-naphthalimide (Easton, Gulbis, Hoskins, Scharfbillig & Tiekink, 1992), but formation of the quinazoline ring leads to a substantial loosening of the N1—C11 bond [1.449 (3) in ($1a$) and 1.439 (3) Å in ($1b$) in comparison with 1.394 (1) Å in (2)], whereas its equivalent, the N1—C12 bond, remains constant [1.401 (3) and 1.399 (3) Å *versus* 1.399 (1) Å]. This may be of importance to the mechanism of hydrolysis of (1).

Experimental

Compounds (1) and (2) were obtained as described earlier (Ponomarev, Skuratova, Lindeman, Sinichkin, Vinogradova & Rusanov, 1994). (1) was crystallized from dimethylformamide; (2) from formic acid.

Compound (1)

Crystal data

C₁₉H₁₀N₂O₂

$M_r = 298.29$

Mo $K\alpha$ radiation

$\lambda = 0.71069$ Å

Triclinic

$P\bar{1}$

$a = 9.065$ (2) Å

$b = 10.858$ (2) Å

$c = 14.118$ (3) Å

$\alpha = 89.59$ (3)°

$\beta = 71.94$ (3)°

$\gamma = 83.63$ (3)°

$V = 1312.4$ (5) Å³

$Z = 4$

$D_x = 1.510$ Mg m⁻³

Cell parameters from 24 reflections

$\theta = 10\text{--}11^\circ$

$\mu = 0.100$ mm⁻¹

$T = 173$ (2) K

Prism

$0.5 \times 0.3 \times 0.25$ mm

Yellow

Data collection

Siemens P3 diffractometer

$\omega/2\theta$ scans

Absorption correction:

none

5811 measured reflections

5810 independent reflections

3772 observed reflections

$[I > 2\sigma(I)]$

$R_{\text{int}} = 0.191$

$\theta_{\text{max}} = 28.97^\circ$

$h = 0 \rightarrow 9$

$k = -14 \rightarrow 14$

$l = -17 \rightarrow 18$

2 standard reflections

monitored every 98

reflections

intensity decay: 2.5%

Refinement

Refinement on F^2

$R[F^2 > 2\sigma(F^2)] = 0.0696$

$wR(F^2) = 0.1679$

$S = 1.134$

5805 reflections

495 parameters

H atoms refined isotropically

$w = 1/[\sigma^2(F_o^2) + (0.1255P)^2 + 0.1195P]$

where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{\text{max}} = 0.003$

$\Delta\rho_{\text{max}} = 0.572$ e Å⁻³

$\Delta\rho_{\text{min}} = -0.392$ e Å⁻³

Extinction correction: none

Atomic scattering factors from *International Tables for Crystallography* (1992, Vol. C, Tables 4.2.6.8 and 6.1.1.4)

Table 1. Fractional atomic coordinates and equivalent isotropic displacement parameters (Å²) for (1)

	$U_{\text{eq}} = (1/3)\sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$			
	<i>x</i>	<i>y</i>	<i>z</i>	U_{eq}
O1	0.3067 (2)	0.09524 (15)	-0.4100 (2)	0.0472 (5)
N1	0.0458 (2)	0.06307 (15)	-0.36653 (13)	0.0231 (4)
N2	-0.2044 (2)	0.02515 (15)	-0.37377 (13)	0.0237 (4)
C1	-0.0115 (2)	-0.1467 (2)	-0.39405 (14)	0.0222 (4)
C2	-0.1150 (3)	-0.2259 (2)	-0.4030 (2)	0.0260 (5)
C3	-0.0694 (3)	-0.3540 (2)	-0.4128 (2)	0.0298 (5)
C4	0.0771 (3)	-0.4026 (2)	-0.4142 (2)	0.0312 (5)
C5	0.3435 (3)	-0.3682 (2)	-0.4131 (2)	0.0331 (5)
C6	0.4483 (3)	-0.2886 (2)	-0.4094 (2)	0.0353 (6)
C7	0.4034 (3)	-0.1606 (2)	-0.4010 (2)	0.0322 (5)
C8	0.2531 (2)	-0.1140 (2)	-0.3949 (2)	0.0261 (5)
C9	0.1880 (3)	-0.3239 (2)	-0.4080 (2)	0.0276 (5)
C10	0.1421 (2)	-0.1945 (2)	-0.39816 (14)	0.0236 (4)
C11	0.2109 (3)	0.0207 (2)	-0.3912 (2)	0.0298 (5)
C12	-0.0619 (2)	-0.0139 (2)	-0.37832 (14)	0.0211 (4)
O1'	0.0752 (2)	0.2488 (2)	-0.3064 (2)	0.0472 (5)
C1'	-0.1636 (2)	0.2315 (2)	-0.33544 (15)	0.0247 (5)
C2'	-0.2568 (2)	0.1494 (2)	-0.35534 (14)	0.0231 (4)
C3'	-0.4093 (3)	0.1912 (2)	-0.3543 (2)	0.0272 (5)
C4'	-0.4657 (3)	0.3135 (2)	-0.3333 (2)	0.0319 (5)
C5'	-0.3736 (3)	0.3957 (2)	-0.3131 (2)	0.0334 (5)
C6'	-0.2240 (3)	0.3551 (2)	-0.3139 (2)	0.0299 (5)
C7'	-0.0058 (3)	0.1888 (2)	-0.3345 (2)	0.0283 (5)
O1a	0.4834 (2)	0.20825 (15)	0.21748 (13)	0.0377 (4)
N1a	0.2854 (2)	0.19098 (15)	0.15327 (13)	0.0248 (4)

N2a	0.0439 (2)	0.1451 (2)	0.13639 (13)	0.0276 (4)
C1a	0.2465 (3)	−0.0197 (2)	0.11641 (15)	0.0255 (5)
C2a	0.1590 (3)	−0.0994 (2)	0.0904 (2)	0.0308 (5)
C3a	0.2205 (3)	−0.2220 (2)	0.0652 (2)	0.0355 (6)
C4a	0.3686 (3)	−0.2649 (2)	0.0651 (2)	0.0347 (6)
C5a	0.6157 (3)	−0.2253 (2)	0.0927 (2)	0.0353 (6)
C6a	0.7014 (3)	−0.1473 (2)	0.1203 (2)	0.0381 (6)
C7a	0.6380 (3)	−0.0258 (2)	0.1485 (2)	0.0334 (5)
C8a	0.4902 (2)	0.0166 (2)	0.1458 (2)	0.0271 (5)
C9a	0.4620 (3)	−0.1862 (2)	0.0907 (2)	0.0303 (5)
C10a	0.3998 (3)	−0.0619 (2)	0.11701 (14)	0.0266 (5)
C11a	0.4253 (3)	0.1451 (2)	0.1760 (2)	0.0272 (5)
C12a	0.1846 (2)	0.1106 (2)	0.13731 (15)	0.0248 (5)
O1'a	0.3434 (2)	0.39510 (14)	0.14197 (14)	0.0421 (5)
C1'a	0.0861 (3)	0.3599 (2)	0.1510 (2)	0.0288 (5)
C2'a	−0.0110 (3)	0.2701 (2)	0.1477 (2)	0.0267 (5)
C3'a	−0.1666 (3)	0.3069 (2)	0.1524 (2)	0.0326 (5)
C4'a	−0.2218 (3)	0.4304 (2)	0.1602 (2)	0.0384 (6)
C5'a	−0.1252 (3)	0.5200 (2)	0.1640 (2)	0.0414 (6)
C6'a	0.0271 (3)	0.4850 (2)	0.1587 (2)	0.0362 (6)
C7'a	0.2471 (3)	0.3233 (2)	0.1487 (2)	0.0295 (5)

Table 2. Selected geometric parameters (Å, °) for (1)

O1—C11	1.221 (3)	O1a—C11a	1.169 (3)
N1—C12	1.401 (3)	N1a—C12a	1.399 (3)
N1—C7'	1.422 (2)	N1a—C11a	1.439 (3)
N1—C11	1.449 (3)	N1a—C7'a	1.446 (3)
N2—C12	1.296 (3)	N2a—C12a	1.293 (3)
N2—C2'	1.378 (2)	N2a—C2'a	1.385 (3)
C1—C12	1.460 (3)	C1a—C12a	1.460 (3)
C8—C11	1.467 (3)	C8a—C11a	1.464 (3)
O1'—C7'	1.188 (3)	O1'a—C7'a	1.216 (3)
C1'—C7'	1.458 (3)	C1'a—C7'a	1.459 (3)
C12—N1—C7'	119.5 (2)	C12a—N1a—C11a	121.4 (2)
C12—N1—C11	122.0 (2)	C12a—N1a—C7'a	119.4 (2)
C7'—N1—C11	118.4 (2)	C11a—N1a—C7'a	119.1 (2)
C12—N2—C2'	119.3 (2)	C12a—N2a—C2'a	119.0 (2)
C2—C1—C10	119.8 (2)	C2a—C1a—C10a	119.9 (2)
C2—C1—C12	119.7 (2)	C2a—C1a—C12a	119.3 (2)
C10—C1—C12	120.5 (2)	C10a—C1a—C12a	120.6 (2)
C7—C8—C10	120.6 (2)	C7a—C8a—C10a	120.9 (2)
C7—C8—C11	119.4 (2)	C7a—C8a—C11a	119.9 (2)
C10—C8—C11	119.9 (2)	C10a—C8a—C11a	119.2 (2)
C1—C10—C8	120.7 (2)	C8a—C10a—C1a	121.2 (2)
O1—C11—N1	120.3 (2)	O1a—C11a—N1a	120.9 (2)
O1—C11—C8	123.5 (2)	O1a—C11a—C8a	121.9 (2)
N1—C11—C8	116.3 (2)	N1a—C11a—C8a	117.3 (2)
N2—C12—N1	124.0 (2)	N2a—C12a—N1a	124.3 (2)
N2—C12—C1	118.1 (2)	N2a—C12a—C1a	118.7 (2)
N1—C12—C1	117.9 (2)	N1a—C12a—C1a	117.0 (2)
C2'—C1'—C7'	120.6 (2)	C2'a—C1'a—C7'a	120.3 (2)
C6'—C1'—C7'	120.5 (2)	C6'a—C1'a—C7'a	120.2 (2)
N2—C2'—C1'	121.2 (2)	N2a—C2'a—C1'a	121.5 (2)
N2—C2'—C3'	118.8 (2)	N2a—C2'a—C3'a	119.0 (2)
O1'—C7'—N1	120.3 (2)	O1'a—C7'a—N1a	121.7 (2)
O1'—C7'—C1'	125.2 (2)	O1'a—C7'a—C1'a	124.5 (2)
N1—C7'—C1'	114.4 (2)	N1a—C7'a—C1'a	113.8 (2)

Compound (2)

Crystal data

C₁₉H₁₂N₂O₃M_r = 316.31

Monoclinic

P2₁/c

a = 9.354 (2) Å

b = 14.130 (3) Å

c = 11.923 (2) Å

β = 111.28 (3)°

V = 1468.4 (5) Å³

Z = 4

D_x = 1.431 Mg m^{−3}

Mo Kα radiation

λ = 0.71069 Å

Cell parameters from 24 reflections

θ = 10–11°

μ = 0.099 mm^{−1}

T = 173 (2) K

Prism

0.5 × 0.4 × 0.35 mm

Yellow

Data collection

Siemens P3 diffractometer

ω/2θ scans

Absorption correction:

none

5391 measured reflections

5132 independent reflections

3967 observed reflections

[I > 2σ(I)]

R_{int} = 0.012

Refinement

Refinement on F²R[F² > 2σ(F²)] = 0.0475wR(F²) = 0.1194

S = 1.049

5126 reflections

265 parameters

H atoms refined isotropically

w = 1/[σ²(F_o²) + (0.0705P)² + 0.4423P]where P = (F_o² + 2F_c²)/3θ_{max} = 35.06°

h = 0 → 13

k = 0 → 22

l = −19 → 17

2 standard reflections

monitored every 98

reflections

intensity decay: 2.7%

(Δ/σ)_{max} = 0.003Δρ_{max} = 0.498 e Å^{−3}Δρ_{min} = −0.201 e Å^{−3}

Extinction correction: none

Atomic scattering factors

from International Tables

for Crystallography (1992,

Vol. C, Tables 4.2.6.8 and 6.1.1.4)

Table 3. Fractional atomic coordinates and equivalent isotropic displacement parameters (Å²) for (2)

	$U_{eq} = (1/3)\sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i \cdot \mathbf{a}_j$		
	x	y	z
O1	0.76656 (11)	−0.03540 (8)	0.29891 (9)
O2	0.97954 (10)	0.13342 (7)	0.07635 (8)
N1	0.87226 (10)	0.05097 (7)	0.18882 (8)
C1	0.71021 (13)	0.15024 (8)	0.02347 (10)
C2	0.69214 (15)	0.20842 (9)	−0.07381 (11)
C3	0.5455 (2)	0.24371 (11)	−0.14309 (13)
C4	0.4204 (2)	0.22262 (10)	−0.11415 (13)
C5	0.30892 (14)	0.13750 (10)	0.01793 (13)
C6	0.32633 (14)	0.07640 (10)	0.11088 (14)
C7	0.47099 (14)	0.03727 (9)	0.17626 (12)
C8	0.59680 (12)	0.06213 (8)	0.14816 (10)
C9	0.43501 (13)	0.16293 (9)	−0.01500 (11)
C10	0.58193 (12)	0.12545 (8)	0.05319 (10)
C11	0.74773 (13)	0.02145 (8)	0.21783 (10)
C12	0.86378 (12)	0.11319 (8)	0.09533 (10)
O1'	0.97394 (13)	0.21800 (7)	0.32657 (9)
N1'	1.06258 (14)	0.17028 (9)	0.52013 (10)
C1'	1.10306 (12)	0.06917 (8)	0.37163 (10)
C2'	1.02245 (12)	0.01878 (7)	0.26692 (9)
C3'	1.07862 (14)	−0.06540 (9)	0.23987 (11)
C4'	1.21895 (15)	−0.10010 (9)	0.31768 (12)
C5'	1.30281 (14)	−0.04983 (9)	0.41986 (12)
C6'	1.24509 (13)	0.03433 (9)	0.44686 (11)
C7'	1.04014 (14)	0.15870 (8)	0.40362 (11)

Table 4. Selected geometric parameters (Å, °) for (2)

O1—C11	1.2195 (14)	C1—C12	1.476 (2)
O2—C12	1.2179 (14)	C8—C11	1.470 (2)
N1—C11	1.3940 (14)	O1'—C7'	1.2315 (15)
N1—C12	1.3991 (14)	N1'—C7'	1.338 (2)
N1—C2'	1.4485 (14)	C1'—C7'	1.502 (2)
C11—N1—C12	125.09 (9)	N1—C11—C8	117.41 (10)
C11—N1—C2'	116.70 (9)	O2—C12—N1	119.89 (10)
C12—N1—C2'	118.09 (9)	O2—C12—C1	123.54 (10)
C2—C1—C10	120.12 (10)	N1—C12—C1	116.57 (10)
C2—C1—C12	119.70 (11)	C6'—C1'—C7'	120.22 (10)
C10—C1—C12	120.18 (10)	C2'—C1'—C7'	121.48 (10)
C7—C8—C10	120.80 (11)	C3'—C2'—N1	118.78 (10)
C7—C8—C11	119.44 (11)	C1'—C2'—N1	119.84 (10)
C10—C8—C11	119.76 (10)	O1'—C7'—N1'	122.58 (11)

C8—C10—C1	120.90 (10)	O1'—C7'—C1'	121.40 (11)
O1—C11—N1	120.03 (10)	N1'—C7'—C1'	116.02 (10)
O1—C11—C8	122.56 (11)		

Refinement was performed on F^2 for all reflections except for five for structure (1) and six for structure (2) with very negative F^2 or flagged for potential systematic errors (e.g. extinction).

For both compounds, program(s) used to solve structures: *SHELXS86* (Sheldrick, 1985); program(s) used to refine structures: *SHELXL93* (Sheldrick, 1993).

Lists of structure factors, anisotropic displacement parameters, H-atom coordinates and bond distances and angles involving non-H atoms have been deposited with the IUCr (Reference: AB1270). Copies may be obtained through The Managing Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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