IMIDAZOQUINAZOLINODIONES - NEW RESULTS

Katarzyna Kieć-Kononowicz, Wolfgang Holzer, and Kurt Mereiter

1 Department of Chemical Technology of Drugs, Collegium Medicum of Jagiellonian University, PL-30688 Kraków, Medyczna 9, Poland

2 Institute of Pharmaceutical Chemistry, University of Vienna, A-1090 Wien, Althanstraße 14, Austria

3 Institute of Mineralogy, Crystallography and Structural Chemistry, Technical University of Vienna, A-1060 Wien, Getreidemarkt 9, Austria

Abstract- X-Ray crystal structure determinations and spectral analyses (NMR, MS) shed new light on the reaction of 5-(Z)-arylidene-2-methylthioimidazolin-4-one (5b) with anthranilic acid, in which apart from the expected 2-(Z)-1-arylidene-2,3,5,10-tetrahydroimidazo[2,1-b]quinazoline-3,5-dione (6b) a new condensed heterocyclic system was identified, i.e. 2-(Z)-arylidene-1,2,4,5-tetrahydroimidazo[1,2-α]quinazoline-1,5-dione (8b). Reaction of the sodium salt of the 4-chloro-substituted arylidene derivative (6a) with methyl iodide surprisingly afforded 2-[(Z)-1-(4-chlorophenyl)methylene]-3-methyl-1,2,3,5-tetrahydroimidazo[1,2-α]-quinazoline-1,5-dione (15).

As a part of our studies on searching for compounds with anticonvulsant properties the known antiepileptic drug phenytoin (5,5-diphenylhydantoin) was modified. Thus, bicyclic and tricyclic derivatives of 5,5-diphenyl- and 5-arylidene-2-thiohydantoins with general structures (1 - 4) were obtained (Scheme 1). Recently, the possible mechanism of interaction of these hydantoins with the benzodiazepine binding site of the GABA_A receptor has attracted our attention. In this context the annelated structures (4), derivatives of 5-arylidene-2-thiohydantoin, seemed the most interesting. The synthesis of such compounds was already described by one of us. Thus (Scheme 2), S-methylated derivatives of 5-arylidene-2-thiohydantoins (5a, 5b) reacted with anthranilic acid (ANTA) to give the tricyclic structures of type (4). However, differences in the reaction behavior of 5a and 5b were observed. In the reaction of 5a only one product (6a) was separated while in the case of 5b two products were
obtained. Based on elemental analysis and $^1$H-NMR and MS spectroscopy these products were assigned as the $Z$ and $E$ diastereomers (6b and 7b)

\[\text{Scheme 1}\]

\[\text{Scheme 2}\]

At that time despite repeated efforts an X-Ray crystal structure analysis was not feasible. With the recent availability of advanced X-Ray diffraction equipment, the successful crystal structure determination of the reaction products of 5b with ANTA permitted a revision of the results towards structures (6b), and, instead of 7b, towards 8b (Scheme 4).

**RESULTS AND DISCUSSION**

Crystal Structures of 2-[(Z)-1-phenylmethylene]-2,3,5,10-tetrahydroimidazo[2,1-b]quinazoline-3,5-dione (6b) and 2-[(Z)-1-phenylmethylene]-1,2,4,5-tetrahydroimidazo[1,2-a]quinazoline-1,5-dione (8b)

The crystal structures of the two compounds were clarified by X-Ray single crystal methods in order to obtain a safe base for their constitution and stereochemistry. Both compounds were recrystallized from dimethyl sulfoxide (DMSO) yielding the corresponding crystalline solvates. While crystals of 6b-dmso were suitable for X-Ray work, those of 8b-dmso were soft and rather labile. Therefore 8b was recrystallized from dimethylformamide (DME) and yielded stable yellow prisms of 8b-dmf which were subsequently studied by X-Ray diffraction. X-Ray structure analysis allowed to confirm the expected structure of 6b and allowed to ascribe the proper structure of the second compound as 8b. Technical details on the crystal structure determinations are described in the experimental section. Views of the molecules including the solvents as encountered in the
crystalline state are shown in Figures 1 and 2. Salient atomic parameters are given in Tables 1 and 2, selected bond lengths and bond angles are compiled in Table 3.

Figure 1. Thermal ellipsoid plot (20% ellipsoids) of C$_{17}$H$_{11}$N$_{5}$O$_{2}$:(CH$_{3}$)$_{2}$SO (6b·dms).  

![Image of Crystal Structure](image1)

Figure 2. Thermal ellipsoid plots (20% ellipsoids) of C$_{17}$H$_{11}$N$_{5}$O$_{2}$:HCON(CH$_{3}$)$_{2}$ (8b·dmf).  

![Image of Crystal Structure](image2)

The benzylideneimidazoquinazolinedione moieties of the two compounds are flat and show only r.m.s. deviations from common planes of 0.094 Å for 6b (0.036 Å without benzylidene moiety), and 0.045 Å for 8b. Thus, the nitrogen atoms are in sp$^2$-hybridized. The bond lengths in both compounds are consistent with the chemical structure diagrams (Figure 3, Scheme 4). The heterocyclic rings show some resonance effects in their C-C and C-N bond lengths, but the C(8)-N(3) bonds are in both compounds consistently the shortest ring bonds. This situation differs distinctly from the related Z-1-ethyl-3-(4-chlorobenzylidene)-1,2,3,5-tetrahydroimidazo[2,1-b]quinazoline-2,5-dione compound of type (3) which contains a formal C-N double bond corresponding to C(8)-N(1) of 6b.

Both 6b·dms and 8b·dmf show clearcut and straight N-H...O hydrogen bonds from the imidazoquinazoline nitrogen N(1) to the solvent oxygen atoms. Both hydrogen bonds are astonishingly similar in size (Table 3) and can be regarded to contribute much to the good stability of the two solvate structures. However, the spatial arrangement of the constituents of the two compounds differs distinctly from one another: 6b·dms exhibits parallel to (100) a pronounced sheet-like arrangement of its constituents, whereas 8b·dmf may be visualized as showing a herringbone-like spatial arrangement of the constituents.
In order to explain the formation of quinazolinodione (8b) the reaction of 5a and 5b with a stoichiometric amount of ANTA was repeated. Additionally, similar reactions were carried out with substrates (5) containing para- dimethylamino-, methoxy- and nitro-substituents (Sc, Sd and Se, respectively). After refluxing the adducts in acetic acid for 4-5 h the separated solids were once recrystallized from DMF (5a-Sc, Se) or acetic acid (5d). Based on ¹H-NMR spectra it was concluded that the course of the reactions...
was different. Linear products (6) were obtained in predominance starting with 5a and 5e. In case of 5e a shorter reaction time (5 h instead of 10 h) probably led to an intermediate and not to the cyclic acid structure. The prolonged time of the reaction (10 h) only, allowed to obtain the cyclic products (6e and 8e) (3:1). In the case of unsubstituted 5b - or its equivalents substituted with electron-donating groups (5c and 5d) - both types of products were obtained in almost equal proportions. Table 4 presents a summary of the reaction times and product ratios obtained.

Table 3. Selected bond lengths [Å] and angles [°] for \( C_{9}H_{7}N_{2}O_{2}(CH_{3})_{2}S \) (6b-dmso) and \( C_{9}H_{7}N_{2}O_{2}HCON(CH_{3})_{2} \) (8b-dmso).

<table>
<thead>
<tr>
<th>Bond lengths [Å]</th>
<th>6b-dmso</th>
<th>8b-dmso</th>
</tr>
</thead>
<tbody>
<tr>
<td>C(1)-C(7)</td>
<td>1.213(3)</td>
<td>1.218(2)</td>
</tr>
<tr>
<td>C(2)-C(9)</td>
<td>1.208(3)</td>
<td>1.210(2)</td>
</tr>
<tr>
<td>N(1)-C(7)</td>
<td>1.389(3)</td>
<td>1.375(2)</td>
</tr>
<tr>
<td>N(1)-C(8)</td>
<td>1.342(3)</td>
<td>1.351(2)</td>
</tr>
<tr>
<td>N(2)-C(7)</td>
<td>1.408(3)</td>
<td>1.389(2)</td>
</tr>
<tr>
<td>N(2)-C(1)</td>
<td>1.408(3)</td>
<td>1.410(2)</td>
</tr>
<tr>
<td>N(2)-C(8)</td>
<td>1.421(3)</td>
<td>1.397(2)</td>
</tr>
<tr>
<td>N(3)-C(8)</td>
<td>1.298(3)</td>
<td>1.299(2)</td>
</tr>
<tr>
<td>N(3)-C(10)</td>
<td>1.403(3)</td>
<td>1.394(2)</td>
</tr>
<tr>
<td>C(1)-C(6)</td>
<td>1.405(3)</td>
<td>1.484(2)</td>
</tr>
<tr>
<td>C(6)-C(7)</td>
<td>1.467(3)</td>
<td>1.484(2)</td>
</tr>
<tr>
<td>C(9)-C(10)</td>
<td>1.487(3)</td>
<td>1.344(3)</td>
</tr>
<tr>
<td>C(10)-C(11)</td>
<td>1.452(3)</td>
<td>1.342(2)</td>
</tr>
<tr>
<td>C(11)-C(12)</td>
<td>1.385(3)</td>
<td>1.383(2)</td>
</tr>
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</table>

In order to explain the formation of 6 and 8 two possible reaction mechanisms were considered. They are based on the observation described in the literature\(^{11-13}\) that the generation of unexpected products in the reaction of hydantoins with compounds containing amino groups could result from the amino group attack on the CO group of the hydantoin ring, its opening and recyclization, as shown below (Scheme 3).

**Scheme 3**
Thus (Scheme 4), formation of 6 is clear through intermediate (9). Formation of 8 could proceed via 9 and the nucleophilic attack of the amino group of ANTA on the carbonyl group, which proceeds intramolecular, leading to the rearrangement product (10) and subsequent cyclization giving product (8). The alternative route of the reaction begins with nucleophilic attack of the second ANTA molecule on 6, followed by ring opening [11], rearrangement to intermediate (12) and elimination of ANTA with formation of 8. For the second reaction pathway speaks the fact that products (8d) and (8e) are obtained in the reaction of 5d and 5e with excess of ANTA. A further support in favor of that pathway is that product (6d) reacted with excess of ANTA yielded 8d. However, methylated 5a as well as quinazolinedione (6a) with excess of ANTA didn’t form significant amounts of 8a.

The proposed reaction mechanism may also explain the formation of the methylated derivatives of para substituted quinazolinediones (15) and (16) (Scheme 5) (especially that of 15, which seems to be an unexpected methylation product (6a)).

Thus, 6a was refluxed in ethanol with a stoichiometric amount of sodium ethoxide in order to obtain its sodium salt. The obtained salt was filtered, dissolved in DMF and alkylated with methyl iodide. In this way product (15) was obtained. Product (15) turned out to be different from 16, the latter obtained as a result of the reaction of 5a with N-methylanthranilic acid. The structures of 15 and 16 were assigned on basis of 1H-NMR chemical shift considerations together with the results of NOE-difference experiments.
NMR Spectroscopic Investigations

The structures of compounds (6a-e, 8a-e, 15, and 16) were confirmed by $^1$H-NMR spectroscopic investigations. Full and unambiguous assignments were achieved on basis of chemical shift and coupling considerations, on NOE-difference experiments\(^\text{14}\) and on 1D TOCSY spectra.\(^\text{15}\) The latter helped to discriminate between spin systems of benzylidene group and the condensed benzene ring of the imidazoquinoline system on the one hand, and - on the other hand - between signals of compounds (6) from those of corresponding compounds (8) in mixtures of both of these species. The signals due to Ph H-2.6 of the (4-substituted) benzylidene system could be easily identified via a strong NOE upon irradiation of the spatially close $=\text{CH}$ singlet (Figure 3).

**Scheme 5**

![Scheme 5](image)

**Figure 3. Important NOEs found with compounds (6, 8, 15, and 16).**

Linear compounds (6) are characterized by an NOE on the signal of H-9 upon perturbation of the NH-resonance, whereas with angular compounds (8) such a through-space connectivity was not observed.
(Figure 3). With compounds (6) and (8) signals of protons H-6 and H-9 are double doublets ($^3J_7-8 \text{ Hz}, \ ^4J_1-15 \text{ Hz}$), whereas H-7 and H-8 have a triplet pattern ($^3J_7-8 \text{ Hz}$) split by a smaller meta coupling ($<1.5 \text{ Hz}$). Both type of compounds show a dynamic behavior in DMSO-$d_6$ solution what is reflected in line broadening for nearly all signals. This phenomenon was also more pronounced observed in the $^{13}$C-NMR spectra of 6b and 8b, which made it impossible to give a full and unambiguous assignment of carbon resonances. In comparison with linear compounds (6), angular systems of type (8) show larger chemical shifts for the ylidene-H (6: 6.71 - 6.78 ppm; 8: 6.93 - 7.03 ppm) as well as for the signals of H-6 - H-9. The marked downfield shift of the signal due to H-9 (δ 8.53 - 8.62 ppm) in angular compounds (8) can be explained by the anisotropy of bond magnetic susceptibility of C=O, coming spatially close to H-9. The chemical shifts of H-6 - H-9 in N-methyl compound (15) assign an angular structure similar to 8, however, a marked NOE on Ph H-2,6 upon irradiation of the NMe resonance hints that the methyl group is attached to N-3 and not to N-4. Attachment of the methyl group at N-10 in the linear compound (16) is confirmed by a strong NOE on H-9 upon perturbation of the NMe signal. The most important NOEs found with 6, 8, 15, and 16 are depicted in Figure 3, also showing the numbering of atoms used in the description of NMR spectra.

**EXPERIMENTAL**

Melting points are uncorrected. TLC was performed using Al. sheets 0.2 mm layer silica gel (60F$_{254}$ Merck); solvent system CHCl$_3$ : AcOEt 1:1. IR were obtained (KBr disc) on Jasco FT IR instrument. UV spectra were measured on V-530 UV/VIS spectrometer. All NMR spectra were recorded on a Varian Unityplus spectrometer (299.95 MHz for $^1$H) from DMSO-$d_6$ solutions at 28°C.

**General procedure of the reaction of 5 with ANTA**

A mixture of 5 (10 mmol) and ANTA (1.4 g, 10 mmol) in acetic acid (20 mL) was refluxed for 4-10 h (Table 4). On the next day the solid was filtered off and recrystallized from an appropriate solvent.

**Reaction of 5a with ANTA**

Raw solid contained 6a and 8a (4:1). The raw solid once crystallized from DMF contained 6a, 1.6 g (yield 50%), mp 322-324 °C (lit., 322-324 °C).

6a: $^1$H-NMR: δ (ppm) 6.73 (s, 1H, ylidene H), 7.22 (m, 1H, H-7), 7.23 (m, 1H, H-9), 7.48 (m, 2H, Ph H-3,5), 7.69 (m, 1H, H-8), 7.97 (m, 1H, H-6), 8.10 (m, 2H, Ph H-2,6), 12.40 (br s, 1H, NH).

8a: $^1$H-NMR: δ (ppm) 6.98 (s, 1H, ylidene H), 7.43 (m, 1H, H-7), 7.51 (m, 2H, Ph H-3,5), 7.82 (m, 1H, H-8), 8.05 (m, 1H, H-6), 8.19 (m, 2H, Ph H-2,6), 8.56 (m, 1H, H-9), 12.40 (br s, 1H, NH).

**Reaction of 5b with ANTA**

Raw solid contained 6b and 8b (1:1). Upon one crystallization of the raw solid from DMF, the ratio of
6b : 8b (1:1), was not changed. The raw solid 2.3 g (yield 79%) was fractionally recrystallized from acetic acid and then DMF giving:

2-[(Z)-1-Phenylmethylene]-2,3,5,10-tetrahydroimidazo[2,1-b]quinazoline-3,5-dione (6b):
0.87 g (yield 30%) mp 305-307 °C (lit., 7 305-307 °C). \( R_f = 0.73 \). \( ^1H\)-NMR: \( \delta \) (ppm) 6.73 (s, 1H, ylidene H), 7.22 (m, 1H, H-7), 7.23 (m, 1H, H-9), 7.32 (m, 1H, Ph H-4), 7.42 (m, 2H, Ph H-3,5), 7.68 (m, 1H, H-8), 7.97 (m, 1H, H-6). 8.05 (m, 2H, Ph H-2,6). 12.23 (br s, 1H, NH).

2-[(Z)-1-Phenylmethylene]-1,2,4,5-tetrahydroimidazo[1,2-a]quinazoline-1,5-dione (8b):
1.04 g (yield 36%), mp 287-289 °C (lit., 7 287-289 °C). \( R_f = 0.78 \). \( ^1H\)-NMR: \( \delta \) (ppm) 6.95 (s, 1H, ylidene H), 7.35 (m, 1H, Ph H-4), 7.41 (m, 1H, H-7), 7.42 (m, 2H, Ph H-3,5), 7.78 (m, 1H, H-8), 8.03 (m, 2H, Ph H-2,6), 8.13 (m, 2H, Ph H-2,6), 8.53 (m, 1H, H-9), 12.60 (br s, 1H, NH).

Reaction of 5c with ANTA
The raw solid was once crystallized from DMF. Yield 54%. The resulting solid contained 6c : 8c (1.2 : 1).

6c column chromatography, eluent CH₂Cl₂, red crystals. 1.16 g (yield 35%), mp 285-287 °C. \( R_f = 0.63 \). \( ^1H\)-NMR: \( \delta \) (ppm) 2.98 (s, 6H, NMe₂), 6.72 (s, 1H, ylidene H), 6.72 (m, 2H, Ph H-3,5), 7.18 (m, 1H, H-7), 7.22 (m, 1H, H-9), 7.65 (m, 1H, Ph H-4), 7.79 (m, 2H, Ph H-2,6), 7.96 (m, 1H, H-6). 12.15 (br s, 1H, NH). Anal Calcd for C₁₉H₁₆N₄O₂: C, 68.66; H, 4.85; N, 16.86. Found: C, 68.65; H, 4.71; N, 16.53.

8c: 0.99 g (yield 30%), \( R_f = 0.72 \). \( ^1H\)-NMR: \( \delta \) (ppm) 2.99 (s, 6H, NMe₂), 7.40 (m, 1H, H-7), 7.79 (m, 1H, H-8), 8.01 (m, 2H, Ph H-2,6), 8.04 (m, 1H, H-6). 8.62 (m, 2H, H-9), 12.15 (br s, 1H, NH).

Reaction of 5d with ANTA
The obtained raw solid once crystallized from acetic acid contained 6d : 8d (1:1).

6d: Yield 35 %, mp 285-286 °C (acetic acid), \( R_f = 0.69 \). \( ^1H\)-NMR: \( \delta \) (ppm) 3.80 (s, 3H, OMe), 6.74 (s, 1H, ylidene H), 6.99 (m, 2H, Ph H-3,5), 7.20 (m, 1H, H-7), 7.22 (m, 1H, H-9), 7.67 (m, 1H, H-8), 7.97 (m, 1H, H-6). 8.03 (m, 2H, Ph H-2,6), 12.30 (br s, 1H, NH). Anal Calcd for C₁₈H₁₃N₃O₃: C, 67.70; H, 4.10; N, 13.16. Found: C, 67.69; H, 3.90; N, 13.04.

8d: Method a: The mixture of 5d (1.24 g, 5 mmol) and ANTA (1.40 g, 10 mmol) in acetic acid (10 mL) was refluxed for 7 h. The solid obtained was recrystallized from acetic acid. It contained 6d : 8d (19:100). 1.29 g (yield 81 %). Method b: The mixture of 6d: (1.60 g, 5 mmol) and ANTA (0.70 g, 5 mmol) in acetic acid (10 mL) was refluxed for 5 h then 0.70 g (5 mmol) of ANTA was added and the mixture was refluxed additionally 5 h. The solid obtained was recrystallized from acetic acid. 1.27 g (yield 80 %).

mp 288-291 °C, \( R_f = 0.74 \). \( ^1H\)-NMR: \( \delta \) (ppm) 3.81 (s, 3H, OMe), 6.97 (s, 1H, ylidene H), 7.01 (m, 2H, Ph H-3,5), 7.41 (m, 1H, H-7), 7.80 (m, 1H, H-8), 8.04 (m, 1H, H-6), 8.14 (m, 2H, Ph H-2,6), 8.58 (m, 1H, H-9), 12.30 (br s, 1H, NH). Anal Calcd for C₁₈H₁₃N₃O₃: C, 67.70; H, 4.10; N, 13.16. Found: C,
67.50; H, 3.96; N, 12.92.

**Reaction of 5e with ANTA**

The reaction mixture was refluxed for 5 h. The solid which appeared was checked by means of TLC ($R_f = 0.00$). The heating was continued for 10 h. The resulting solid was once crystallized from DMF. It contained 6e : 8e (3:1).

6e: $R_f = 0.70$ (in ammonia atmosphere the orange spot becomes quickly almost black, the colour disappears after few seconds). mp 329-332 °C (DMF). $^1$H-NMR: $\delta$ (ppm) 6.78 (s, 1H, ylidene H), 7.24 (m, 1H, H-7), 7.25 (m, 1H, H-9), 7.70 (m, 1H, H-8), 7.98 (m, 1H, H-6), 8.24 (m, 2H, Ph H-3,5), 8.32 (m, 2H, Ph H-2,6), 12.65 (br s, 1H, NH). UV-VIS (DMF): 349 nm (log $\varepsilon = 3.33$), 435 nm (log $\varepsilon = 2.92$), 599 nm (log $\varepsilon = 3.54$). IR: 3434, 3247, 1761, 1694, 1669. $\beta$(C=O), 1616, 1581, 1511, 1335, 763. Anal. Calcd for C$_{17}$H$_{11}$N$_3$O$_4$: C, 61.08; H, 3.01; N, 16.76. Found: C, 61.05; H, 2.78; N, 16.43.

8e: obtained as 8d - Method a: Reaction time 12 h (in ammonia atmosphere the yellow spot remains yellow). Yield 78 %, mp 343-345 °C (DMF). $^1$H-NMR: $\delta$ (ppm) 7.03 (s, 1H, ylidene H), 7.43 (m, 1H, H-7), 7.82 (m, 1H, H-8), 8.05 (m, 1H, H-6), 8.26 (m, 2H, Ph H-3,5), 8.38 (m, 2H, Ph H-2,6), 8.53 (m, 1H, H-9), 12.65 (br s, 1H, NH). UV-VIS (DMF): 332 nm (log $\varepsilon = 4.01$), 418 nm (log $\varepsilon = 3.94$), 511 nm (log $\varepsilon = 3.73$). IR: 3432, 1733, 1697 (C=O), 1648, 1564, 1483, 1168, 760. Anal. Calcd for C$_{17}$H$_{10}$N$_4$O$_4$: C, 61.08; H, 3.01; N, 16.76. Found: C, 61.17; H, 2.96, N, 16.54.

2-[(Z)-1-(4-Chlorophenyl)methylene]-3-methyl-1,2,3,5-tetrahydroimidazo[1,2-a]quinazoline-1,5-dione (15)
Described in literature as 1-methyl-Z-2-(4-chlorobenzylidene)-2,3,4,5-tetrahydro[2,1-b]quinazoline-3,5-dione. mp 297-298 °C (lit., 296-298 °C). $^1$H-NMR: $\delta$ (ppm) 3.54 (s, 3H, 3-Me), 6.82 (s, 1H, ylidene H), 7.47 (m, 1H, H-7), 7.54 (m, 2H, Ph H-3,5), 7.86 (m, 1H, H-8), 8.12 (m, 1H, H-6), 8.27 (m, 2H, Ph H-2,6), 8.61 (m, 1H, H-9). IR: 1716, 1686 (C=O), 1648, 1568, 1548, 760, 748.

2-[(Z)-1-(4-Chlorophenyl)methylene]-10-methyl-2,3,5,10-tetrahydroimidazo[2,1-b]quinazoline-3,5-dione (16)
Described in literature as 1-methyl-Z-2-(4-chlorobenzylidene)-2,3,4,5-tetrahydro[2,1-b]quinazoline-3,5-dione. mp 316-317 °C (lit., 315-317 °C). $^1$H-NMR: $\delta$ (ppm) 3.74 (s, 3H, 3-Me), 6.82 (s, 1H, ylidene H), 7.33 (m, 1H, H-7), 7.50 (m, 2H, Ph H-3,5), 7.51 (m, 1H, H-9), 7.82 (m, 1H, H-8), 8.08 (m, 1H, H-6), 8.22 (m, 2H, Ph H-2,6). IR: 1780, 1756 (C=O), 1648, 1568, 1548, 760, 748.

**X-Ray Structure Determination of 2-[(Z)-1-Phenylmethylene]-2,3,5,10-tetrahydroimidazo[2,1-b]quinazoline-3,5-dione dimethyl sulfoxide solvate (6b.dmso)**

Crystal data: C$_{19}$H$_{17}$N$_3$O$_4$S = C$_{19}$H$_{17}$N$_3$O$_4$·(CH$_3$)$_2$SO, $M_r = 367.42$, monoclinic, space group $P2_1/c$ (No. 14). $a = 6.849(3)$ Å, $b = 11.834(4)$ Å, $c = 21.868(8)$ Å, $\beta = 93.15(1)^\circ$, $V = 1769.7(12)$ Å$^3$, $Z = 4$, $D_x = 1.379$ g cm$^{-3}$, $\lambda = 0.71073$ Å, $\mu = 0.207$ mm$^{-1}$, $T = 301K$. A yellow plate, 0.50 x 0.32 x 0.05 mm, crystallized from hot dimethyl sulfoxide was used for data collection with a Siemens Smart CCD
X-Ray Structure Determination of 2-[(Z)-1-Phenylmethylene]-1,2,4,5-tetrahydroimidazo[1,2-a]quinazoline-1,5-dione dimethylformamide solvate (8b.dmf)

Crystal data: C_{17}H_{11}N_{3}O_{2}.HCON(CH_{3})_{2} = C_{29}H_{18}N_{4}O_{3}, M, = 362.38, monoclinic, space group P2_{1}/c (No. 14), a = 6.353(2) Å, b = 18.495(5) Å, c = 15.437(5) Å, β = 98.10(2)°, V = 1795.7(9) Å^{3}, Z = 4, D_{x} = 1.340 g cm^{-3}, λ = 0.71073 Å, μ = 0.093 mm^{-1}, T = 301 K. A yellow prism, 0.6 x 0.2 x 0.2 mm, crystallized from dimethylformamide at rt was used for data collection with a Siemens Smart CCD diffractometer. Intensity data with θ ≤ 25° were collected over more than one hemisphere of the reciprocal space using 0.3° ω-scan frames. They were corrected for Lp, decay, absorption and related effects (correction factors 0.57 - 0.96); 11718 reflections collected, 3163 independent, R_{int} = 0.040. Structure solution with direct methods, structure refinement with SHELXL93. Hydrogen atoms were located from a difference Fourier map and were refined riding with the atoms to which they were bonded. The final refinement varied 248 parameters and used all 3163 independent reflections weighted by w=1/[σ^{2}(F_{o}^{2})+(0.051P)^{2}+0.08P]. Final R1 = 0.071, wR2 = 0.107 and S = 1.01 for all data, R1 = 0.039 for the 2090 reflections with F_{o}^{2} > 2σ(F_{o}^{2}). Excursions in final difference Fourier map between -0.129 and 0.155 e Å^{-3}. Atomic coordinates are presented in Table 2.\textsuperscript{17}  

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REFERENCES AND NOTES

9. The crystal structure of 8b.dmso was determined, but the results were not satisfying according to present standards: Orthorhombic, s.g. Pbn21, a = 4.995(1), b = 22.290(4), c = 32.280(6) Å, Z = 8 C19H17NiO3S, pseudo-centrosymmetric structure with N-H...O hydrogen bonds between pyrimidine N and dmso.
17. Further details of the crystal structure investigations are available from the 'Fachinformationszentrum Karlsruhe', D-76344 Eggenstein-Leopoldshafen (Germany), on quoting the depository number CSD-410576 (6b dmso), and CSD-410577 (8b dmf).

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