A new polycyclic system containing the 1,4-benzodiazepine and isoindolinone fragments: synthesis and structure

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Dehydration of 2-(3,3-dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]-diazepin-11-yl)benzoic acid (**3a**) and 2-(3,3-dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)-5,6-dimethoxybenzoic acid (**3b**) in boiling acetic or propionic anhydride gave 7,7-dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo[2,3:5,6][1,4]diazepino[7,1-*a*]-isoindole-5,15-dione (**5a**) and 1,2-dimethoxy-7,7-dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo-[2,3:5,6][1,4]diazepino[7,1-*a*]isoindole-5,15-dione (**5b**), respectively. The products obtained are representatives of a novel fused pentacyclic system. The structures of compounds **3b** and **5a** were unambiguously proved by X-ray diffraction.

Key words: 3-(2-aminophenylamino)-5,5-dimethylcyclohex-2-en-1-one, 2-formylbenzoic acid, 2-formyl-5,6-dimethoxybenzoic (opianic) acid, hexahydrodibenzo[*b*,*e*][1,4]diazepines, hexahydrodibenzo[*b*,*e*]isoindolino[3,2-*g*][1,4]diazepines, X-ray diffraction analysis.

Recently,[1—3] we have reported on the synthesis of phthalimidines from arylaminophthalides and *N*-acyl-2-formylbenzohydrazones. The present work is devoted to the use of 2-formylbenzoic acid and 2-formyl-5,6-dimethoxybenzoic (opianic) acid for the synthesis of a novel fused system containing the benzodiazepine and phthalimidine fragments. It is known that 3-(2-aminophenylamino)-5,5-dimethylcyclohex-2-en-1-one (**1**) smoothly enters into acid-catalyzed condensation with aldehydes at room temperature to give tricyclic dibenzo[1,4]diazepines in high yields,[4] which exhibit narcotic and analgesic activities.[5,6]

Using 2-formylbenzoic (**2a**) and opianic acids (**2b**) as the aldehyde component in this reaction, we obtained 2-(3,3-dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)benzoic acid (**3a**) and 2-(3,3-dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)-5,6-dimethoxybenzoic acid (**3b**), respectively (Scheme 1). The presence of the carboxyl function in the starting compounds allowed this synthesis to be performed without adding acid catalysts. In boiling acetic or propionic anhydride, these compounds were easily transformed into 7,7-dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo[2,3:5,6][1,4]diazepino[7,1-*a*]isoindole-5,15-dione (**5a**) and 1,2-dimethoxy-7,7-dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo[2,3:5,6][1,4]diazepino[7,1-*a*]-isoindole-5,15-dione (**5b**), respectively (see Scheme 1). These are derivatives of a novel fused pentacyclic system. Earlier,[4] the easy formation of N(10)-acetyl derivatives has been noted for reactions of analogs of compounds **3a**,**b** with Ac_2O. Apparently, the cyclization **3** → **5** also proceeds through acyl intermediates **4a**,**b**.

Compound **3a** obtained according to a known procedure[4] has been characterized in a study[7] dealing with MS fragmentation of similar dibenzodiazepinones. Its mass spectrum contains a peak corresponding to compound **5a**. Structure **5a** has been described in a patent,[8] though without any mention of its synthesis or properties. Data on compounds **3b** and **5b** are lacking in the literature. Structures **3b** and **5a** were unambiguously proved by X-ray diffraction from crystals grown by slow evaporation of their solutions in acetonitrile.

The general view of structure **5a** is shown in Fig. 1. The central seven-membered ring has a distorted boat conformation. The C(8), N(1), N(2), and C(10) atoms are coplanar. The C(15) and C(20) atoms deviate from their plane by 0.886(4) and 0.822(4) Å, respectively, while the C(9) atom deviates by 0.113(4) Å; the N(1), C(15), C(20),

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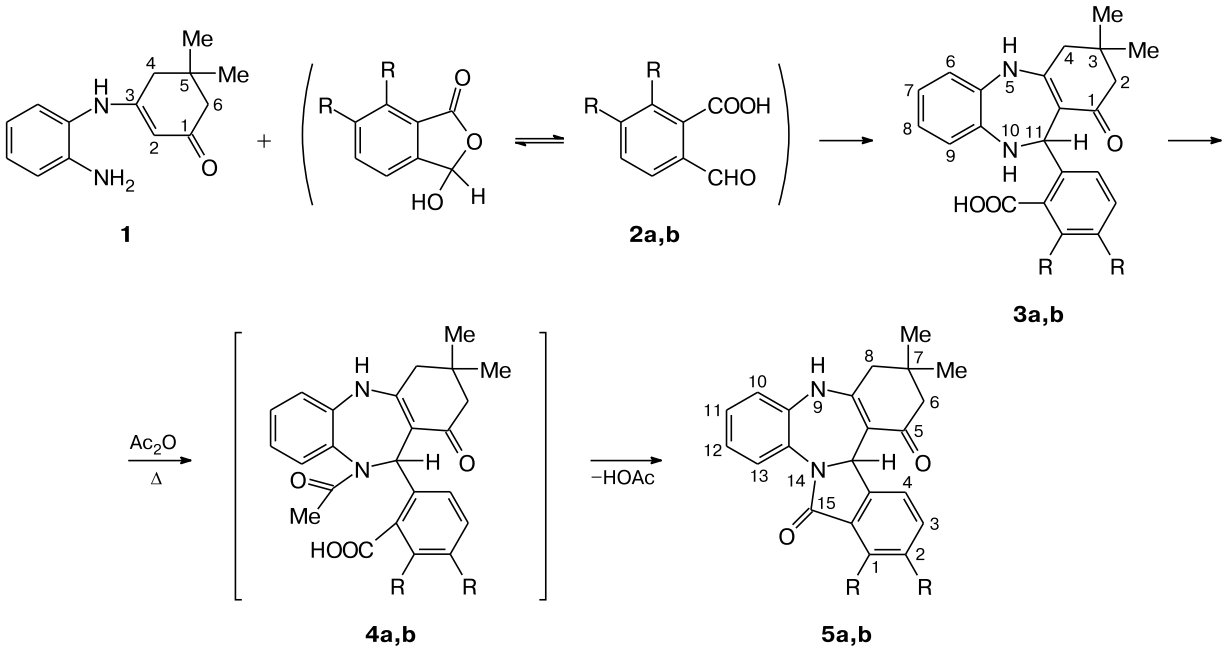
Scheme 1



R = H (**a**), OMe (**b**)

and N(2) atoms are also coplanar. The dimethylcyclohexenone fragment at the C(9) and C(10) atoms has a distorted half-chair conformation, while the other rings are planar. The group N(2)—H(2) is involved in the hydrogen bond N(2)—H(2)...O(1) (H...O, 2.21 Å; N...O, 3.022(2) Å; the angle N—H...O, 149°), thus forming chains along the crystallographic direction [101].

The general view of structure **3b** with a solvate acetonitrile molecule in a ratio of 1 : 1 is shown in Fig. 2. The molecules are linked by the hydrogen bond N(2)—H(2)...N(1S) (H...O, 2.15 Å; N...O, 3.010(5) Å; the angle N—H...O, 160°).

The central fragment in structure **3b** somewhat differs from that in structure **5a**. The seven-membered ring has

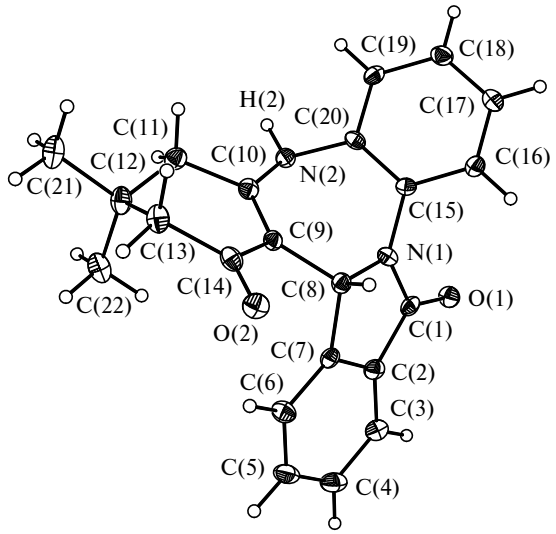


Fig. 1. General view of structure **5a** with atomic thermal displacement ellipsoids (p = 50%).

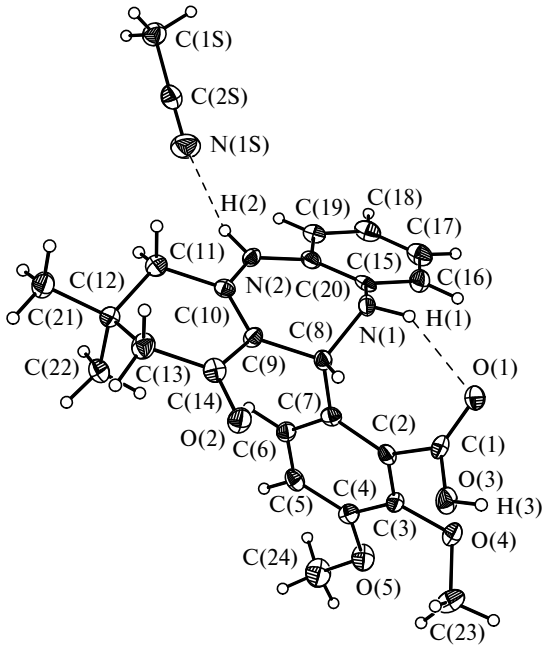


Fig. 2. General view of structure **3b** with atomic thermal displacement ellipsoids (p = 50%).

a skew boat conformation. The N(1), C(15), C(20), and N(2) atoms are coplanar (as in **5a**), while the C(8), N(1), N(2), and C(10) atoms are not. The dimethylcyclohexenone fragment has nearly the same conformation as in **5a**; all the aromatic rings are planar. The carboxy group is far off the plane of the adjacent benzene ring: the torsion angle C(3)—C(2)—C(1)—O(1) is 116.6(4)°. Apparently, the orientation of the carboxy group is due to a cooperative effect of its steric interaction with the nearest methoxy group, the intramolecular hydrogen bond N(1)—H(1)...O(1) (H...O, 2.16 Å; N...O, 2.928(4) Å; the angle N—H...O, 144°), and the strong intermolecular hydrogen bond O(3)—H(3)...O(2) ($-x, 1-y, z-0.5$) (H...O, 1.70 Å; O...O, 2.534(3) Å; the angle O—H...O, 168°), which gives rise to chains along the axis *c*.

The ^{1}H NMR spectra of compounds **3a,b** in DMSO-d_6 are generally similar to the ^{1}H NMR spectrum of their analog containing unsubstituted phenyl at the C(11) atom.[4] As in this analog, one CH_2 group in compounds **3a**,**b** is manifested as a singlet, while the other, as two doublets with J = 16 Hz (the AB system). However, their ^{1}H NMR spectra show specific features because of the presence of the carboxy group in the *ortho*-position of the phenyl substituent. The ^{1}H NMR spectrum of the above phenyl analog shows, as expected, two doublets for the protons at the C(11) and N(10) atoms (see Ref. 4), while the corresponding signals in the spectra of compounds **3a**,**b** are two singlets.

The signals in the ^{1}H NMR spectrum of compound **3a** were assigned from 2D ^{1}H—^{1}H COSY experiment. To assign the signals in the ^{13}C NMR spectra, we used 2D ^{1}H—^{13}C correlation experiments (HSQC and HMBC).

Chemical shifts in the ^{15}N NMR spectrum were determined using ^{1}H—^{15}N HMBC experiment. The data obtained are summarized in Table 1.

The ^{1}H—^{15}N HMBC spectrum shows a cross peak for the N(5) atom as a doublet with $J_{N(5),H(5)}$ = 90 Hz. The absence of cross peaks for the N(10) atom in the 2D ^{1}H—^{15}N HMBC spectrum is probably due to an exchange between the protons of the groups O(8′)H (δ 13.25) and N(10)H (δ 5.56) and the protons of water (δ 3.37) present in the solvent (DMSO-d_6). Because of this, the signals of these protons are broadened in the ^{1}H NMR spectrum. The exchange process was detected using an EXSY experiment (exchange spectroscopy). The EXSY spectrum contain cross peaks for three groups of the protons involved in the exchange: O(8′)H, N(10)H, and H_2O (Fig. 3).

To find out whether compound **3a** behaves like this because of the *ortho*-position of the carboxy group in the phenyl substituent, we synthesized its *para*-analog **3c** (Scheme 2).

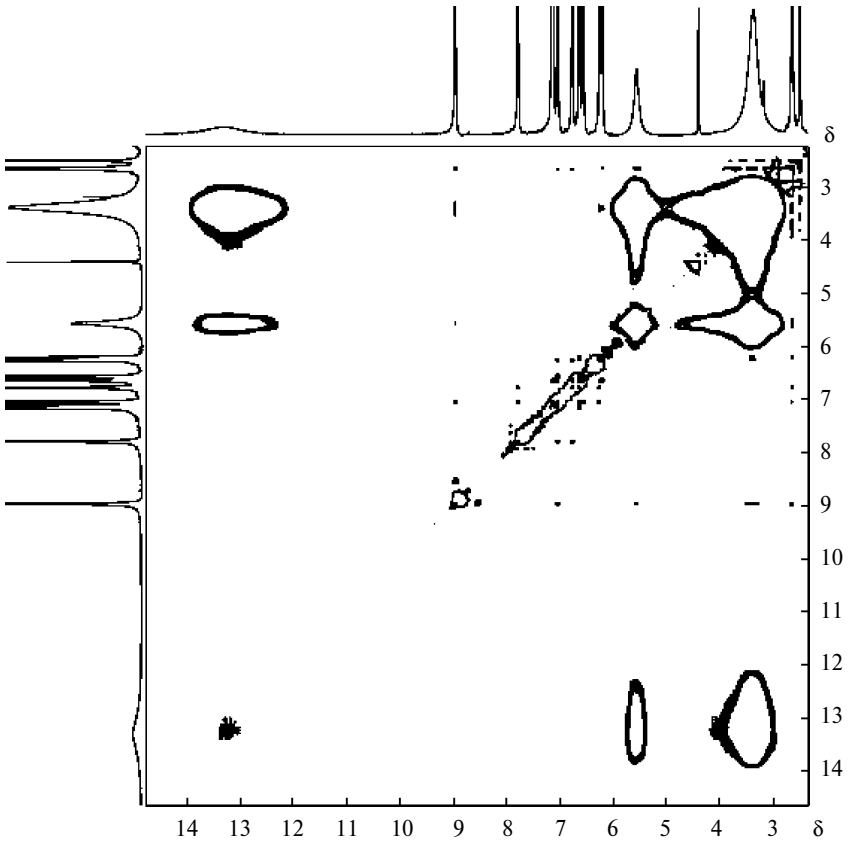
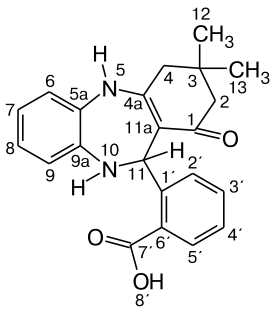


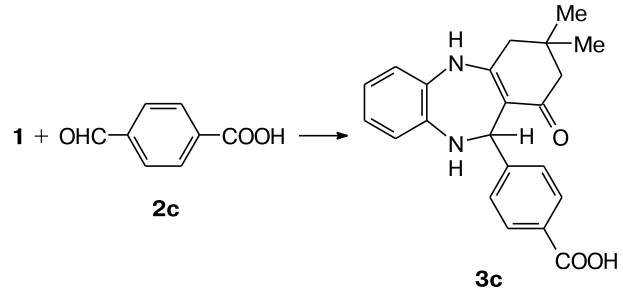
Fig. 3. 2D EXSY spectrum of the intermolecular exchange between the protons of the groups O(8′)H (δ 13.25), N(10)H (δ 5.56), and H_2O (δ 3.37) for compound **3a**.

Table 1. ^{1}H, ^{13}C, and ^{15}N NMR spectra of compound **3a** in DMSO-d_6



No. of the atom	NMR, δ		
	^{1}H	^{13}C	^{15}N
1	—	192.31	—
2	2.09	49.53	—
3	—	31.84	—
4	2.64	44.22	—
4a	—	155.13	—
5	8.96	—	122.0
5a	—	131.39	—
6	7.04	120.11	—
7	6.64	120.37	—
8	6.56	123.03	—
9	6.25	120.63	—
9a	—	137.47	—
10	5.56	—	82.10
11	6.20	54.96	—
11a	—	109.36	—
1′	—	145.09	—
2′	6.77	126.36	—
3′	7.15	131.07	—
4′	7.13	126.38	—
5′	7.79	130.46	—
6′	—	130.03	—
7′	—	169.68	—
8′	13.25	—	—
12	1.08	28.23	—
13	1.01	27.44	—

Scheme 2



As expected, the ^{1}H NMR spectrum of compound **3c** has a common shape with two doublets for the H(10) and H(11) protons. This suggests that the anomalous features in the ^{1}H and ^{15}N NMR spectra of compound **3a** result from prototropic rotation of the carboxy group about the σ-bond to the phenyl substituent, which brings about rapid exchange of the proton at the N(10) atom.

Experimental

IR spectra were recorded on a Varian Excalibur 3100 FT-IR instrument (ATR-FTIR). The ^{1}H NMR spectra of compounds **3b** and **5a**,**b** were recorded on a UNITY-300 spectrometer (Varian). The ^{1}H and ^{13}C NMR spectra of compound **3a** and the ^{1}H NMR spectrum of compound **3c** were recorded on an AVANCE-600 spectrometer (Bruker). Mass spectra were measured on a Finnigan MAT INCOS 50 instrument (direct inlet probe).

2-(3,3-Dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)benzoic acid (3a). A hot solution of 2-formylbenzoic acid **2a** (0.75 g, 5 mmol) in MeOH (5 mL) was added to a hot solution of compound **1** (1.15 g, 5 mmol) in MeOH (5 mL). The mixture was heated to boiling and cooled with ice water, while triturating with a rod. The precipitate that formed was filtered off, washed with cold MeOH, and dried. The yield of compound **3a** was 1.4 g (73%), colorless solid, m.p. 186—188 °C (from $MeNO_2$) (*cf.* Ref. 7: m.p. 179—181 °C). Found (%): C, 72.83; H, 6.21; N, 7.82. $C_{22}H_{22}N_2O_3$. Calculated (%): C, 72.91; H, 6.21; N, 7.73. IR, ν/cm^{-1}: 3884, 3336, 3289 (NH), 1680, 1667, 1624 (CO), 1602, 1573, 1519 (arom.). ^{1}H NMR (DMSO-d_6), δ: 1.01, 1.08 (both s, 6 H, Me); 2.03, 2.16 (both d, 2 H, CH_2, J = 16.0 Hz); 2.62 (s, 2 H, CH_2); 5.56 (s, 1 H, N(10)H); 6.20 (s, 1 H, C(11)H); 6.25 (d, 1 H, CH arom., J = 7.6 Hz); 6.50—7.50 (m, 6 H, CH arom.); 7.79 (d, 1 H, CH arom., J = 7.0 Hz); 8.96 (s, 1 H, N(5)H); 13.50 (br.s, 1 H, OH). MS, *m/z*: 362 $[M]^+$.

2-(3,3-Dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)-5,6-dimethoxybenzoic acid (3b) (solvate with MeCN) was obtained from opianic acid **2b** (0.46 g, 2.5 mmol) in MeCN (3 mL) and compound **1** (0.6 g, 2.6 mmol) in MeCN (3 mL) as described for the synthesis of compound **3a**. Crystallization was promoted by using seed crystals. The yield of compound **3b** was 0.83 g (72%), light yellow solid, m.p. 150—155 °C (foaming). Found (%): C, 67.63; H, 6.25; N, 8.72. $C_{24}H_{26}N_2O_5 \cdot MeCN$ ($C_{26}H_{29}N_3O_5$). Calculated (%): C, 67.37; H, 6.31; N, 9.06. IR, ν/cm^{-1}: 3340, 3312, 3247 (NH), 2253 (CN), 1682, 1630 (CO), 1610, 1572, 1485 (arom.). ^{1}H NMR (DMSO-d_6), δ: 1.05, 1.13 (both s, 6 H, Me); 1.98 (s, 3 H, MeCN); 2.02, 2.13 (both d, 2 H, CH_2, J = 16.0 Hz); 2.59 (s, 2 H, CH_2); 3.69, 3.77 (both s, 6 H, OMe); 4.87 (s, 1 H, N(10)H); 5.58 (s, 1 H, C(11)H); 6.35 (m, 2 H, CH arom.); 6.57 (m, 3 H, CH arom.); 6.96 (m, 1 H, CH arom.); 8.75 (s, 1 H, N(5)H). MS, *m/z*: 422 $[M]^+$.

4-(3,3-Dimethyl-1-oxo-2,3,4,5,10,11-hexahydro-1*H*-dibenzo[*b*,*e*][1,4]diazepin-11-yl)benzoic acid (3c) (solvate with MeOH). A hot solution of compound **1** (1.15 g, 5 mmol) in MeOH (5 mL) was added to a hot solution of 4-formylbenzoic acid (0.75 g, 5 mmol) in MeOH (20 mL). After the reaction mixture was brought to boiling, the formation of a colorless crystalline precipitate was observed. The reaction mixture was refluxed for 1 min and cooled to room temperature. The precipitate was filtered off, washed with MeOH and light petroleum, and dried. The yield of compound **3c** was 1.35 g (75%), m.p. 262—264 °C. Found (%): C, 70.05; H, 6.71; N, 7.25. $C_{22}H_{22}N_2O_3 \cdot MeOH$ ($C_{23}H_{26}N_2O_4$). Calculated (%): C, 70.03; H, 6.64; N, 7.10. IR, ν/cm^{-1}: 3409, 3281, 3238 (OH, NH), 1679 (CO), 1605, 1588, 1503 (arom.). ^{1}H NMR (DMSO-d_6), δ: 1.02, 1.07 (both s, 6 H, Me); 2.08, 2.18 (both d, 2 H, CH_2, J = 16.0 Hz); 2.59 (s, 2 H, CH_2); 3.15 (d, 3 H, Me (MeOH), J = 5.2 Hz); 4.11 (q, 1 H, OH (MeOH), J = 5.2 Hz); 5.72 (d, 1 H, N(10)H, J = 5.5 Hz); 6.25 (d, 1 H, C(11)H, J = 5.5 Hz); 6.50 (dd, 1 H, CH arom., ^3J = 7.4 Hz, ^4J = 2.0 Hz); 6.57 (m, 2 H, CH arom.); 6.92 (dd, 1 H, CH arom., ^3J = 7.6 Hz, ^4J = 1.8 Hz); 7.16 (d, 2 H, C(2′)H, C(6′)H, J = 8.5 Hz); 7.66 (d, 2 H, C(3′)H, C(5′)H, J = 8.5 Hz); 8.83 (s, 1 H, N(5)H); 12.71 (s, 1 H, COOH).

7,7-Dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo[2,3:5,6][1,4]diazepino[7,1-*a*]isoindole-5,15-dione (5a). ***A.*** A mixture of com-

Table 2. Crystallographic parameters and the data collection statistics for compounds **3b** and **5a**

Compound	**3b**	**5a**
Molecular formula	$C_{26}H_{29}N_3O_5$	$C_{26}H_{29}N_3O_5$
Molecular weight	463.52	463.52
Crystal color	Colorless	Colorless
Crystal shape	Plates	Prisms
Crystal dimensions/mm	0.22×0.16×0.02	0.15×0.10×0.10
T/K	100(2)	100(2)
Crystal system	Orthorhombic	Monoclinic
Space group	$Pca2_1$	Pn
a/Å	18.693(5)	6.8450(9)
b/Å	12.136(3)	11.4650(14)
c/Å	11.102(3)	11.4815(15)
β/deg	90	105.486(3)
V/Å^3	2518.5(11)	868.33(19)
Z	4	2
d_{calc}/g cm^{-3}	1.222	1.317
Absorption coefficient μ/mm^{-1}	0.086	0.085
F(000)	984	364
θ scan range/deg	1.68—27.00	1.78—29.00
Number of measured reflections	16655	10124
Number of independent reflections	2891	2313
R_{int}	0.1051	0.0731
Number of parameters refined	312	237
Number of reflections with $I \geq 2\sigma(I)$	2104	1814
Scan completeness (%)	99.8	99.7
GOOF	1.011	1.071
$R_1(F)$* for reflections with $I \geq 2\sigma(I)$)	0.0470	0.0457
$wR_2(F^2)$** for all reflections	0.1025	0.0959
Residual electron density (max/min)/e Å^{-3}	0.276/−0.226	0.255/−0.246

* $R_1 = \Sigma||F_o| - |F_c||/\Sigma(F_o)$.
** $wR_2 = \Sigma[w(F_o^2 - F_c^2)^2]/\Sigma[w(F_o^2)^2]^{1/2}$.

pound **3a** (0.72 g, 2 mmol) and Ac_2O (2.5 mL) was heated to homogenization, refluxed for 1 min, and cooled to room temperature. Then Pr^{i}OH (3 mL) was added and the mixture was cooled with ice water. The precipitate that formed was filtered off, washed with cold Pr^{i}OH and light petroleum, and dried. The yield of compound **5a** was 0.31 g (46%), colorless solid, m.p. 280—284 °C (from MeCN). Found (%): C, 76.94; H, 6.05; N, 8.23. $C_{22}H_{20}N_2O_2$. Calculated (%): C, 76.72; H, 5.85; N, 8.13. IR, ν/cm^{-1}: 3308, 3240 (NH), 1682, 1624 (CO), 1596, 1501 (arom.). ^{1}H NMR ($CDCl_3$), δ: 0.67, 1.01 (both s, 6 H, Me); 1.90—2.50 (m, 4 H, CH_2); 6.10 (s, 1 H, CH); 6.60—7.20 (m, 4 H, NH, CH arom.); 7.38 (dd, 1 H, CH arom., ^3J = 7.8 Hz, ^4J = 1.2 Hz); 7.55, 7.95 (both m, 4 H, CH arom.).

B. Compound **5a** was also obtained in a similar way from compound **3a** (0.34 g, 0.9 mmol) and propionic anhydride (2 mL). The yield was 0.21 g (65%).

1,2-Dimethoxy-7,7-dimethyl-4b,7,8,9-tetrahydro-6*H*-dibenzo[2,3:5,6][1,4]diazepino[7,1-*a*]isoindole-5,15-dione (5b) was obtained from compound **3b** (0.42 g, 1 mmol) and propionic anhydride (2 mL) as described for the synthesis of compound **5a** (the amount of Pr^{i}OH was 5 mL). The yield was 0.17 g (42%), colorless solid, m.p. 242—245 °C (from MeCN). Found (%): C, 71.13; H, 6.21; N, 7.15. $C_{24}H_{24}N_2O_4$. Calculated (%): C, 71.27; H, 5.98; N, 6.93. IR, ν/cm^{-1}: 3284, 3230 (NH), 1678, 1630 (CO), 1611, 1586, 1536, 1511, 1499 (arom.). ^{1}H NMR ($CDCl_3$), δ: 0.71, 0.97 (both s, 6 H, Me); 1.90—2.50 (m, 4 H, CH_2); 3.91, 4.10 (both s, 6 H, OMe); 5.96 (s, 1 H, CH); 6.70—7.10 (m, 5 H, NH, CH arom.); 7.55 (dd, 1 H, CH arom., ^3J = 7.8 Hz, ^4J = 1.2 Hz); 7.59 (d, 1 H, CH arom., J = 8.7 Hz). MS, m/z: 404 $[M]^+$.

X-ray diffraction study of compounds 3b and 5a. Their single crystals suitable for X-ray diffraction were grown by slow evaporation of the corresponding solutions in acetonitrile.

Experimental reflection intensities were measured on a SMART APEX2 CCD diffractometer (λ(Mo-Kα) = 0.71073 Å, graphite monochromator, ω scan mode). The initial intensity reflection arrays were processed with the APEX2 program package.[9] The structures were solved by the direct methods and refined by the full-matrix least-squares method in the anisotropic approximation for the non-hydrogen atoms on F^2_{hkl}. The hydrogen atoms were located geometrically, except for the H atoms at oxygen and nitrogen, which were located from the difference electron-density maps and normalized to distances of 0.85 (for O) and 0.90 Å (for N). All the H atoms were refined using a riding model ($U_{iso}(H) = nU_{eq}(C,O,N)$, where n = 1.5 for the methyl C atoms and the O atoms and n = 1.2 for the other C atoms and the N atoms). The structures were solved and refined with the SHELXTL program.[10] The crystallographic parameters and the data collection statistics are given in Table 2.

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