Chemistry of acyl(imidoyl)ketenes
5.* Synthesis and structure of 2-(2-oxo-2*H*-1,4-benzoxazin-3-yl)-4-(2,4,6-trimethylbenzoyl)-3-(2,4,6-trimethylbenzoyloxy)pyrido[2,1-*c*][1,4]benzoxazine-1,5-dione

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2-Oxo-2*H*-1,4-benzoxazin-3-yl-(2,4,6-trimethylbenzoyl)ketene, which is generated upon thermal decarbonylation of 3-(2,4,6-trimethylbenzoyl)pyrrolo[2,1-*c*][1,4]benzoxazine-1,2,4-trione, undergoes dimerization through [4+2] cycloaddition to give 2-(2-oxo-2*H*-1,4-benzoxazin-3-yl)-4-(2,4,6-trimethylbenzoyl)-3-(2,4,6-trimethylbenzoyloxy)pyrido[2,1-*c*]-[1,4]benzoxazine-1,5-dione. The resulting compound formed a crystal solvate with *p*-xylene, whose crystal and molecular structure was established by X-ray diffraction analysis.

Key words: dimerization, imidoylketene, pyrroledione, crystal and molecular structure.

Acyl(imidoyl)ketenes contain the acyl and imidoyl groups conjugated with the C=C ketene bond, which allows these compounds to enter into different [4+2] cycloaddition reactions either as a dienophile with the participation of a particular multiple bond or as a diene with the participation of the acylketene or imidoylketene fragments. However, intramolecular conversions are preferential for the known types of acyl(imidoyl)ketenes. Intramolecular cyclization of *N*-benzylimidoyl(aroyl)ketenes, which are generated upon thermal decarbonylation of 4,5-diaroyl-1-benzylpyrrole-2,3-diones, is accompanied by redox processes to form substituted furan-2-ones and furoisoquinolinones.[2] *N*-Arylimidoyl(acyl)ketenes undergo intramolecular cyclization to give substituted 4-quinolones. In this case, the direction of the reaction remains unchanged when a substituent is introduced at one of the *ortho* positions of the aromatic ring at the imidoyl nitrogen atom, including a substituent capable of being involved in cycloaddition.[3]

Aroyl-(2-oxo-2*H*-1,4-benzoxazin-3-yl)ketenes, which are generated upon thermal decarbonylation of 3-aroylpyrrolo[2,1-*c*][1,4]benzoxazine-1,2,4-triones,[4] are representatives of *N*-arylimidoyl(acyl)ketenes, in which the aromatic substituent at the imidoyl nitrogen atom is fixed (as a result of its participation in the condensed system) in such a manner that the above-described cyclization to form 4-quinolones is impossible.

Storage of a solution of 3-(2,4,6-trimethylbenzoyl)-pyrrolo[2,1-*c*][1,4]benzoxazine-1,2,4-trione (**1**)[5] in Dowtherm A at 158—160 °C afforded a high-melting compound, which is, according to the data of elemental analysis and mass spectrometry, a dimer of 2-oxo-2*H*-1,4-benzoxazin-3-yl-(2,4,6-trimethylbenzoyl)ketene (**2**). Based on the published data on acyl- and imidoylketenes, the occurrence can be suggested of [4+2] cyclodimerization, in the course of which one molecule acts as a dienophile through the C=C bond of the ketene fragment and the second molecule acts as a diene through the acylketene (pathway *a*)[6] or imidoylketene (pathway *b*)[7] fragments. As a result, substituted pyran-2,4-dione **3** or pyrido[2,1-*c*][1,4]benzoxazine-1,3,5-trione **4**, respectively, can be formed. The structure of the dimer could not be unambiguously established based on the spectral data. X-ray diffraction analysis of crystals of the product grown from *p*-xylene allowed us to identify the resulting compound as 2-(2-oxo-2*H*-1,4-benzoxazin-3-yl)-4-(2,4,6-trimethylbenzoyl)-3-(2,4,6-trimethylbenzoyloxy)pyrido[2,1-*c*]-[1,4]benzoxazine-1,5-dione (**5**) (Scheme 1).

Apparently, the molecule of ketene **2** is involved in [4+2] cyclodimerization as a diene through the imidoylketene fragment (pathway *b*). In this case, the resulting dimer **4** undergoes 1,3-acylotropic rearrangement[8] to form compound **5**.

The structure of molecule **5** is shown in Fig. 1. The bond lengths and bond angles are given in Tables 1 and 2, respectively. The C(18)—C(19) (1.352 Å),

* For Part 4, see Ref. 1.
† Deceased.

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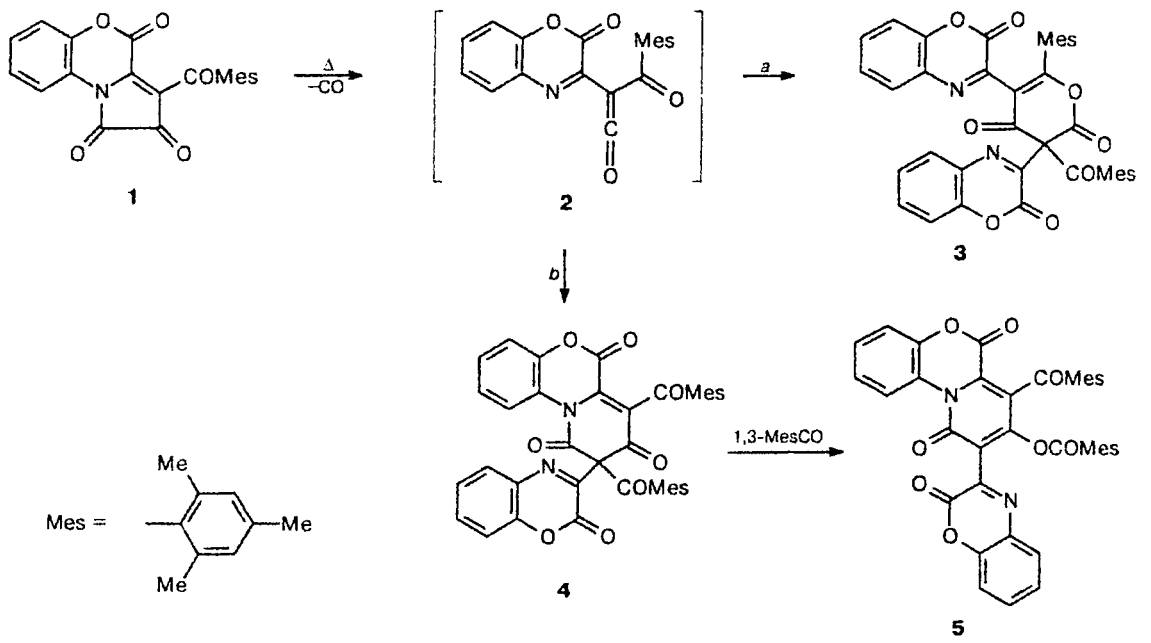
Scheme 1



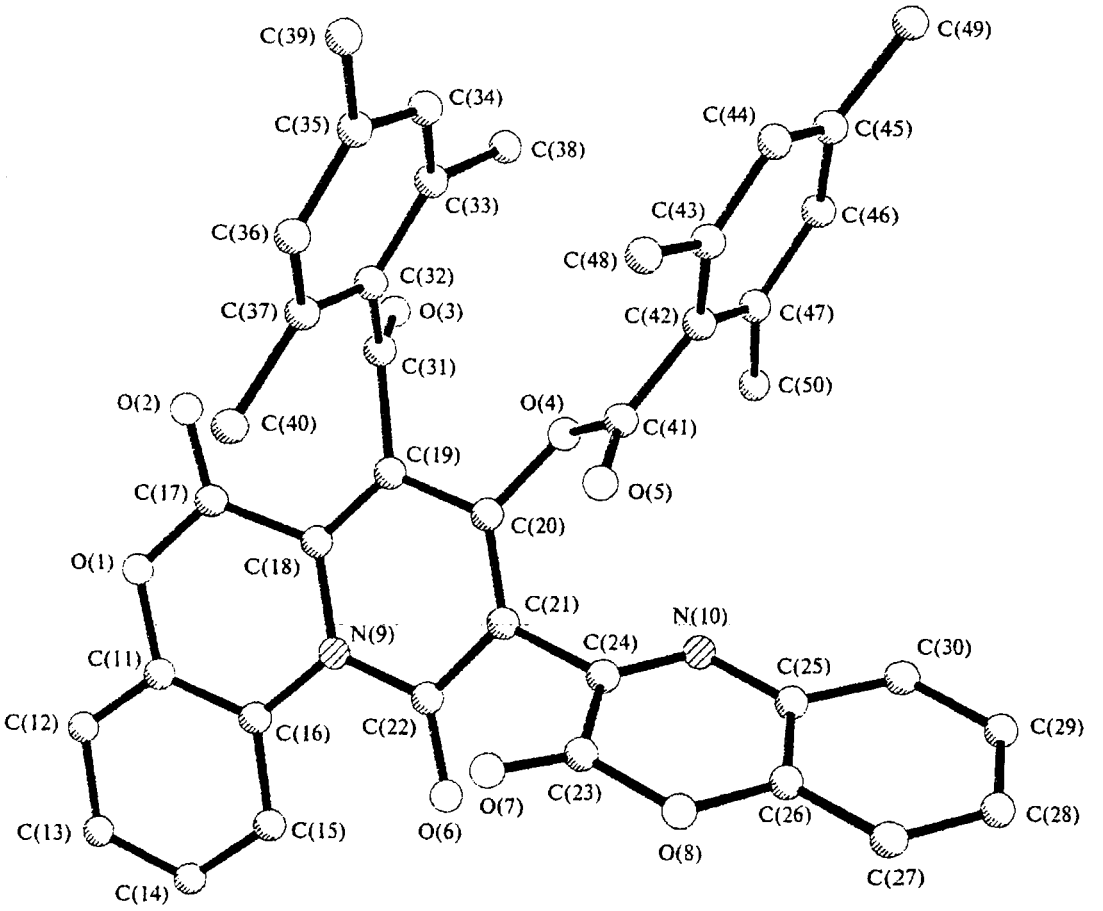


Fig. 1. Structure of molecule 5.

Table 1. Bond lengths (d) in molecules 5

Bond	d/Å	Bond	d/Å	Bond	d/Å	Bond	d/Å
O(1)—C(17)	1.364(5)	C(11)—C(16)	1.371(6)	C(26)—C(27)	1.373(6)	C(42)—C(47)	1.406(6)
O(1)—C(11)	1.390(5)	C(11)—C(12)	1.380(6)	C(27)—C(28)	1.364(8)	C(43)—C(44)	1.386(6)
O(2)—C(17)	1.185(5)	C(12)—C(13)	1.370(7)	C(28)—C(29)	1.377(8)	C(43)—C(48)	1.502(7)
O(3)—C(31)	1.214(5)	C(13)—C(14)	1.363(7)	C(29)—C(30)	1.380(7)	C(44)—C(45)	1.369(7)
O(4)—C(41)	1.377(5)	C(14)—C(15)	1.399(6)	C(31)—C(32)	1.480(6)	C(45)—C(46)	1.379(6)
O(4)—C(20)	1.382(4)	C(15)—C(16)	1.402(6)	C(32)—C(37)	1.408(5)	C(45)—C(49)	1.507(7)
O(5)—C(41)	1.206(5)	C(17)—C(18)	1.478(6)	C(32)—C(33)	1.418(5)	C(46)—C(47)	1.393(6)
O(6)—C(22)	1.240(5)	C(18)—C(19)	1.352(5)	C(33)—C(34)	1.384(6)	C(47)—C(50)	1.512(7)
O(7)—C(23)	1.204(5)	C(19)—C(20)	1.421(5)	C(33)—C(38)	1.490(6)	C(51)—C(52)	1.368(8)
O(8)—C(23)	1.365(5)	C(19)—C(31)	1.536(6)	C(34)—C(35)	1.375(6)	C(51)—C(56)	1.379(9)
O(8)—C(26)	1.392(5)	C(20)—C(21)	1.349(5)	C(35)—C(36)	1.372(6)	C(51)—C(58)	1.499(10)
N(9)—C(18)	1.400(5)	C(21)—C(22)	1.436(5)	C(35)—C(39)	1.509(7)	C(52)—C(53)	1.371(9)
N(9)—C(22)	1.404(5)	C(21)—C(24)	1.478(6)	C(36)—C(37)	1.391(6)	C(53)—C(54)	1.362(8)
N(9)—C(16)	1.442(5)	C(23)—C(24)	1.484(6)	C(37)—C(40)	1.529(6)	C(54)—C(55)	1.385(9)
N(10)—C(24)	1.273(5)	C(25)—C(26)	1.365(6)	C(41)—C(42)	1.481(6)	C(54)—C(57)	1.495(11)
N(10)—C(25)	1.403(5)	C(25)—C(30)	1.395(6)	C(42)—C(43)	1.392(6)	C(55)—C(56)	1.382(9)

Table 2. Bond angles (ω) in molecule 5

Angle	ω/deg	Angle	ω/deg	Angle	ω/deg	Angle	ω/deg
C(17)—O(1)—C(11)	121.1(4)	C(18)—C(19)—C(31)	125.7(4)	C(27)—C(28)—C(29)	120.5(5)	C(43)—C(42)—C(47)	121.1(4)
C(41)—O(4)—C(20)	122.9(3)	C(20)—C(19)—C(31)	115.4(4)	C(28)—C(29)—C(30)	119.9(6)	C(43)—C(42)—C(41)	118.2(4)
C(23)—O(8)—C(26)	120.0(4)	C(21)—C(20)—O(4)	125.4(4)	C(29)—C(30)—C(25)	119.9(5)	C(47)—C(42)—C(41)	120.6(4)
C(18)—N(9)—C(22)	119.9(3)	C(21)—C(20)—C(19)	121.3(4)	O(3)—C(31)—C(32)	123.2(4)	C(44)—C(43)—C(42)	118.0(5)
C(18)—N(9)—C(16)	118.3(3)	O(4)—C(20)—C(19)	113.2(4)	O(3)—C(31)—C(19)	116.4(4)	C(44)—C(43)—C(48)	119.0(5)
C(22)—N(9)—C(16)	121.8(4)	C(20)—C(21)—C(22)	119.8(4)	C(32)—C(31)—C(19)	119.7(4)	C(42)—C(43)—C(48)	123.0(4)
C(24)—N(10)—C(25)	117.9(4)	C(20)—C(21)—C(24)	124.3(4)	C(37)—C(32)—C(33)	118.8(4)	C(45)—C(44)—C(43)	122.4(5)
C(16)—C(11)—C(12)	123.6(5)	C(22)—C(21)—C(24)	115.9(4)	C(37)—C(32)—C(31)	123.2(4)	C(44)—C(45)—C(46)	118.9(5)
C(16)—C(11)—O(1)	122.5(4)	O(6)—C(22)—N(9)	121.7(4)	C(33)—C(32)—C(31)	118.0(4)	C(44)—C(45)—C(49)	121.3(5)
C(12)—C(11)—O(1)	114.0(4)	O(6)—C(22)—C(21)	120.1(4)	C(34)—C(33)—C(32)	118.7(4)	C(46)—C(45)—C(49)	119.8(6)
C(13)—C(12)—C(11)	119.3(5)	N(9)—C(22)—C(21)	118.1(4)	C(34)—C(33)—C(38)	117.7(4)	C(45)—C(46)—C(47)	121.6(5)
C(14)—C(13)—C(12)	119.3(5)	O(7)—C(23)—O(8)	118.3(5)	C(32)—C(33)—C(38)	123.5(4)	C(46)—C(47)—C(42)	117.9(4)
C(13)—C(14)—C(15)	121.4(5)	O(7)—C(23)—C(24)	125.7(5)	C(35)—C(34)—C(33)	122.8(4)	C(46)—C(47)—C(50)	117.6(5)
C(14)—C(15)—C(16)	119.9(5)	O(8)—C(23)—C(24)	115.9(4)	C(36)—C(35)—C(34)	118.1(4)	C(42)—C(47)—C(50)	124.5(4)
C(11)—C(16)—C(15)	116.5(4)	N(10)—C(24)—C(21)	120.6(4)	C(36)—C(35)—C(39)	120.8(5)	C(52)—C(51)—C(56)	116.2(7)
C(11)—C(16)—N(9)	119.0(4)	N(10)—C(24)—C(23)	124.0(4)	C(34)—C(35)—C(39)	121.1(5)	C(52)—C(51)—C(58)	123.2(8)
C(15)—C(16)—N(9)	124.5(4)	C(21)—C(24)—C(23)	115.3(4)	C(35)—C(36)—C(37)	122.2(5)	C(56)—C(51)—C(58)	120.6(8)
O(2)—C(17)—O(1)	117.7(4)	C(26)—C(25)—C(30)	118.5(4)	C(36)—C(37)—C(32)	119.3(4)	C(51)—C(52)—C(53)	121.8(7)
O(2)—C(17)—C(18)	124.1(4)	C(26)—C(25)—N(10)	121.7(4)	C(36)—C(37)—C(40)	116.6(4)	C(54)—C(53)—C(52)	122.6(7)
O(1)—C(17)—C(18)	117.8(4)	C(25)—C(26)—C(27)	121.9(5)	C(32)—C(37)—C(40)	124.1(4)	C(53)—C(54)—C(55)	116.2(7)
C(19)—C(18)—N(9)	121.5(4)	C(25)—C(26)—O(8)	120.1(4)	O(5)—C(41)—O(4)	121.4(4)	C(55)—C(54)—C(57)	122.5(7)
C(19)—C(18)—C(17)	118.1(4)	C(27)—C(26)—O(8)	118.0(5)	O(5)—C(41)—C(42)	128.8(4)	C(56)—C(55)—C(54)	121.1(7)
N(9)—C(18)—C(17)	120.2(4)	C(28)—C(27)—C(26)	119.3(6)	O(4)—C(41)—C(42)	109.8(4)	C(51)—C(56)—C(55)	121.9(7)
C(18)—C(19)—C(20)	119.0(4)						

C(20)—C(21) (1.349 Å), and N(10)—C(24) (1.273 Å) distances correspond to the double bonds. The remaining bonds also have standard values. Hence, the molecule exists in the form with the localized double bonds without noticeable conjugation between the fragments. The oxazine ring of the tricyclic pyridobenzoxazine fragment is nonplanar. The folding angle along the O(1)...N(9) line is 171.5°. At the same time, there is, apparently, the C(15)—H(15)...O(6) intramolecular hydrogen bond. The C(15)...O(6) and H(15)...O(6) distances are 2.65 and 2.0 Å, respectively. The angle at the hydrogen atom is 124°. Apparently, this fact is responsible for an elongation of the C(22)=O(6) bond (1.240 Å) compared to the remaining four bonds in the carbonyl groups (1.185–1.214 Å). The presence of the C(15)—H(15)...O(6) hydrogen bond is also evidenced by the low-field position (at δ 9.05) of the signal of this hydrogen atom in the ^{1}H NMR spectra. The angle between the planar benzoxazine group at the C(21) atom and the mean plane of the tricyclic fragment is 120°. The C(22)C(21)C(24)C(23) torsion angle is −62.7°. The C(31)=O(3) and C(41)=O(5) carbonyl

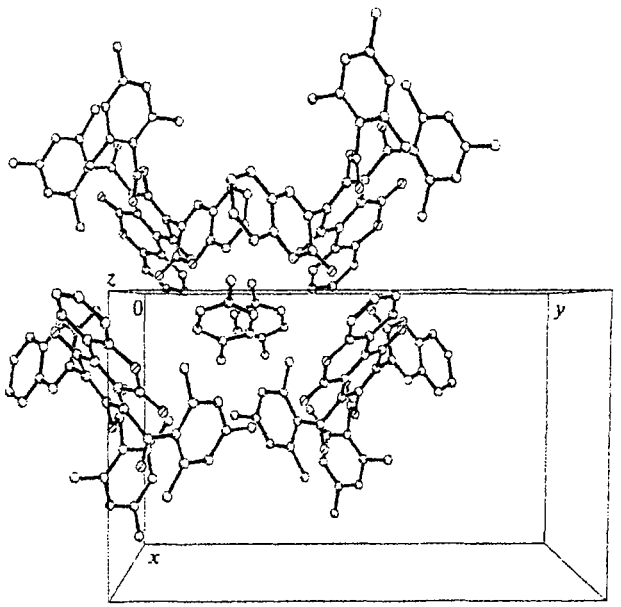


Fig. 2. Fragment of the crystal structure of **5**.

groups are noncoplanar with the planes of the corresponding trimethylbenzene groups. The O(3)C(31)C(32)C(33) and O(5)C(41)C(42)C(43) torsion angles are −31.0° and −39°, respectively. The orientations of the trimethylbenzoyl group and the trimethylbenzoyloxy group with respect to the pyridobenzoxazine fragment are characterized by C(18)C(19)C(31)C(32) and C(19)C(20)O(4)C(41) torsion angles of 114° and −51°, respectively. The O(3) and O(5) atoms are in *trans* orientations with respect to the plane passing through the C(31), C(19), C(20), and O(4) atoms. The planes of the trimethylbenzene groups are parallel to each other. In the crystal, there are no shortened intermolecular contacts. The molecular packing is such that channels are formed along the *c* axis of the crystal. The channels are occupied by *p*-xylene molecules (Fig. 2).

Experimental

The IR spectrum of the synthesized compound was recorded on a UR-20 instrument. The ^{1}H NMR spectrum was obtained on an RYa-2310 spectrometer (60 MHz, HMDS as the internal standard). The mass spectrum (EI) was measured on an MKh-1320 instrument (ionizing voltage was 70 eV). The reaction was performed in Dowtherm A, which is a eutectic mixture of biphenyl and diphenyl ether.[9]

2-(2-Oxo-2*H*-1,4-benzoxazin-3-yl)-4-(2,4,6-trimethylbenzoyl)-3-(2,4,6-trimethylbenzoyloxy)pyrido[2,1-*c*][1,4]-benzoxazine-1,5-dione (5). A solution of compound **1** (1.44 g, 0.04 mol) in Dowtherm A (10 mL) was kept at 158—160 °C for

Table 3. Coordinates of nonhydrogen atoms (×10^4) and their equivalent isotropic thermal parameters ($U_{eq} \cdot 10^3$/Å^2) in the structure of **5**

Atom	*x*	*y*	*z*	U_{eq}
O(1)	7566(3)	−593(2)	−643(2)	49(1)
O(2)	6138(3)	−1086(2)	−94(2)	66(1)
O(3)	4276(3)	−601(2)	784(2)	51(1)
O(4)	5434(2)	147(1)	2383(2)	41(1)
O(5)	6648(3)	−11(2)	3494(2)	55(1)
O(6)	8647(3)	1188(2)	1309(2)	63(1)
O(7)	9343(3)	548(2)	2901(2)	72(1)
O(8)	8926(3)	1350(2)	3712(2)	56(1)
N(9)	7740(3)	334(2)	606(2)	33(1)
N(10)	6701(3)	1443(2)	2896(2)	42(1)
C(11)	8334(4)	−70(2)	−664(3)	42(1)
C(12)	8984(4)	−67(3)	−1327(3)	49(1)
C(13)	9754(5)	434(3)	−1423(3)	54(2)
C(14)	9852(4)	921(3)	−867(3)	53(1)
C(15)	9215(4)	909(2)	−185(3)	44(1)
C(16)	8426(4)	399(2)	−78(2)	33(1)
C(17)	6783(4)	−633(2)	−70(3)	43(1)
C(18)	6873(4)	−148(2)	586(2)	31(1)
C(19)	6153(3)	−212(2)	1186(2)	31(1)
C(20)	6309(4)	208(2)	1863(2)	34(1)
C(21)	7194(4)	643(2)	1936(2)	33(1)
C(22)	7912(4)	744(2)	1279(2)	39(1)
C(23)	8644(4)	954(3)	3069(3)	47(1)
C(24)	7467(4)	1056(2)	2651(2)	39(1)
C(25)	7002(4)	1808(2)	3592(2)	37(1)
C(26)	8090(5)	1771(2)	3985(3)	47(1)
C(27)	8397(6)	2140(3)	4654(3)	63(2)
C(28)	7590(6)	2551(3)	4942(3)	70(2)
C(29)	6485(6)	2603(3)	4559(3)	63(2)
C(30)	6183(5)	2231(3)	3887(3)	51(1)
C(31)	5158(4)	−712(2)	1213(2)	35(1)
C(32)	5234(4)	−1230(2)	1831(2)	32(1)
C(33)	4204(4)	−1387(2)	2210(2)	35(1)
C(34)	4258(4)	−1872(2)	2788(3)	43(1)
C(35)	5261(4)	−2221(2)	2996(2)	41(1)
C(36)	6254(4)	−2066(2)	2626(3)	43(1)
C(37)	6276(4)	−1567(2)	2064(2)	38(1)
C(38)	3069(5)	−1039(3)	2046(3)	52(1)
C(39)	5276(6)	−2757(3)	3615(3)	62(2)
C(40)	7452(5)	−1433(3)	1721(4)	62(2)
C(41)	5661(4)	49(2)	3196(2)	39(1)
C(42)	4544(4)	−6(2)	3584(2)	35(1)
C(43)	4450(4)	−489(2)	4161(3)	40(1)
C(44)	3374(5)	−573(3)	4475(3)	50(1)
C(45)	2425(4)	−189(3)	4254(3)	52(1)
C(46)	2543(4)	300(3)	3699(3)	49(1)
C(47)	3594(4)	404(2)	3351(2)	41(1)
C(48)	5450(5)	−924(3)	4447(3)	51(1)
C(49)	1282(6)	−271(4)	4633(5)	85(2)
C(50)	3646(5)	965(3)	2768(4)	59(2)
C(51)	9637(6)	−2252(4)	3330(4)	81(2)
C(52)	9206(6)	−2776(3)	3720(4)	75(2)
C(53)	8547(5)	−2702(3)	4365(4)	72(2)
C(54)	8289(5)	−2107(3)	4668(4)	73(2)
C(55)	8684(7)	−1571(3)	4263(5)	88(2)
C(56)	9345(7)	−1646(4)	3610(5)	88(2)
C(57)	7610(10)	−2044(5)	5393(6)	119(3)
C(58)	10371(8)	−2315(5)	2629(6)	124(3)

Table 4 Coordinates of H atoms ($\times 10^4$) in the structure of **5**

Atom	x	y	z	Atom	x	y	z
H(12)	8902(39)	−426(22)	−1687(26)	H(46)	1891(40)	574(22)	3466(26)
H(13)	10123(40)	390(23)	−1928(27)	H(48A)	5795(43)	−1137(23)	3958(29)
H(14)	10362(40)	1311(22)	−894(26)	H(48B)	5078(45)	−1264(25)	4720(28)
H(15)	9295(38)	1244(21)	203(24)	H(48C)	6053(46)	−699(25)	4751(29)
H(27)	9173(43)	2028(25)	4937(29)	H(49A)	1046(61)	−696(33)	4638(42)
H(28)	7748(45)	2858(24)	5425(30)	H(49B)	780(62)	−14(35)	4403(43)
H(29)	5817(42)	2925(24)	4747(27)	H(49C)	1385(57)	−211(33)	5223(39)
H(30)	5418(42)	2241(23)	3664(27)	H(50A)	4320(50)	1182(27)	2789(32)
H(34)	3554(37)	−1997(20)	3076(24)	H(50B)	3035(50)	1236(27)	2830(31)
H(36)	6932(39)	−2243(22)	2787(26)	H(50C)	3512(49)	796(25)	2224(33)
H(38A)	3179(43)	−531(27)	2032(28)	H(52)	9392(48)	−3251(27)	3537(30)
H(38B)	2631(42)	−1137(24)	2554(29)	H(53)	8344(46)	−3103(27)	4643(31)
H(38C)	2592(44)	−1165(24)	1563(29)	H(55)	8559(54)	−1138(31)	4406(35)
H(39A)	6188(47)	−3100(26)	3772(29)	H(56)	9644(51)	−1286(30)	3278(34)
H(39B)	4931(54)	−3102(29)	3460(34)	H(57A)	7464(89)	−1655(45)	5392(58)
H(39C)	5367(51)	−2641(27)	4148(32)	H(57B)	7974(86)	−2153(45)	5864(49)
H(40A)	7347(46)	−1414(27)	1100(32)	H(57C)	6847(77)	−2406(40)	5419(52)
H(40B)	8060(49)	−1782(28)	1924(31)	H(58A)	10569(81)	−1852(41)	2444(52)
H(40C)	7731(49)	−988(28)	1949(33)	H(58B)	10034(80)	−2725(39)	2236(50)
H(44)	3345(39)	−992(22)	4834(26)	H(58C)	11121(78)	−2463(42)	2786(54)

10 min and then cooled. The precipitate that formed was filtered off. The yield was 0.5 g (36%), m.p. 272—274 °C (with decomp., from MeCN). Found (%): C, 72.19; H, 4.46; N, 4.29. $C_{40}H_{30}N_2O_8$. Calculated (%): C, 72.06; H, 4.54; N, 4.20. MS, *m/z*: 666 $[M]^+$. IR, ν/cm^{-1}: 1740 (COO), 1655 (C(1)=O, COC_6H_2), 1598 (C=C, C=N). ^1H NMR (DMSO-d_6), δ: 2.15 (s, 12 H, 4 *o*-CH_3); 2.29 (s, 6 H, 2 *p*-CH_3); 7.19 (m, 11 H, $C_6H_4 + C_6H_3 + 2\ C_6H_2$); 9.05 (m, 1 H, C(10)H).

X-ray diffraction study of compound 5. Crystals suitable for X-ray diffraction study were prepared by recrystallization of compound **5** from *p*-xylene. The identity of the samples recrystallized from MeCN and *p*-xylene was determined by TLC on Silufol UV-254 plates using a 5 : 1 benzene : ether mixture as the eluent. Compound **5** cocrystallized with *p*-xylene in an equimolar ratio. Crystals of $C_{40}H_{30}N_2O_8 \cdot C_6H_4(CH_3)_2$ belong to the monoclinic system: a = 11.452(2), b = 20.516(4), c = 16.701(3) Å, β = 94.42(3)°, V = 3912.2(12) Å^3, M = 772.82, d_{calc} = 1.312 g cm^{-3}, Z = 4, space group $P2_1/c$. The X-ray diffraction data set was collected on an automated four-circle KM-4 diffractometer (KUMA DIFFRACTION) with the χ geometry using the θ—2θ scanning technique and monochromated Cu-Kα radiation in the angle range of 3.4° < θ < 80.2°. A total of 8358 independent reflections were measured. Absorption was ignored (μ = 0.728 mm^{-1}). The structure was solved by the direct statistical method followed by a series of successive electron density maps. The hydrogen atoms were placed in geometrically calculated positions and only their positional parameters were refined. The thermal parameters of the H atoms were fixed and were 1.2 times (1.5 times for the methyl groups) as large as the corresponding values of the carbon atoms. The full-matrix least-squares refinement with anisotropic thermal parameters for nonhydrogen atoms converged to R = 0.042 using 1746 reflections with $I \geq 2\sigma(I)$. The atomic coordinates are given in Tables 3 and 4. (The parameters of anisotropic thermal vibrations can be obtained from the authors.) All calculations were carried out on a PC/AT computer using the SHELX 86[10] and SHELXL 93[11] program packages.

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