

## CYCLOADDITION IN CONDENSED ISOINDOLES

### 1. PREPARATION OF 2-ARYL-3-METHYL-4-OXO- 3,4-DIHYDROQUINAZOLINE

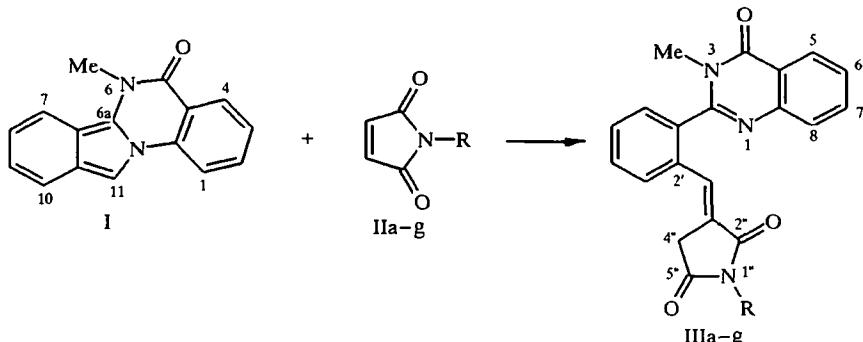
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*Reaction of 6-methyl-5,6-dihydroisoindolo[2,1-*a*]quinazolin-5-one with maleimide derivatives has been studied under conditions of thermodynamic control. X-ray diffraction structural analysis showed that the reaction products are 2-{2-[*(E*)-1-*R*-2,5-dioxopyrrolidinidenemethyl]phenyl}-3-methyl-4-oxo-3,4-dihydroquinazolines. These products are supposed to be formed by rearrangement of the corresponding Diels – Alder adducts formed involving the isoindole part of the molecule.*

Considerable localization of the double bonds in the isoindole fragment is a characteristic feature of 6-methyl-5,6-dihydroisoindolo[2,1-*a*]quinazolin-5-one (I) [1]. Analysis of the static reactivity indices indicated that compound I is most similar to the ancestor isoindole in its capacity to act as a diene in the Diels – Alder reaction [2]. PPP and CNDO/2 calculations showed that cycloaddition should proceed at C<sub>(6a)</sub> and C<sub>(11)</sub> in this system [2]. However, the reaction of I with maleimide derivatives (IIa-g) upon heating in various solvents including CHCl<sub>3</sub>, CH<sub>2</sub>Cl<sub>2</sub>, ethanol, 2-propanol, and toluene unexpectedly gave 2-{[*(E*)-1-*R*-2,5-dioxopyrrolidinidenemethyl]phenyl}-3-methyl-4-oxo-3,4-dihydroquinazolines (IIIa-g). The highest yields of the latter product were achieved by heating the reagents in 2-propanol at reflux for 1 h (in the case of imide IIe) or 2 h (for the other imides) (see Table 1).

The structures of compounds IIIa-g were established by elemental analysis, spectral data, and, in the case of IIIc, by X-ray diffraction analysis.



II, III a R = H, b R = Ph, c R = CH<sub>2</sub>Ph, d R = C<sub>6</sub>H<sub>4</sub>Me-*p*,  
e R = C<sub>6</sub>H<sub>4</sub>NO<sub>2</sub>-*p*, f R = OC<sub>6</sub>H<sub>4</sub>Me-*p*, g R = 2-Npht

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The IR spectra of these products have C=O group stretching bands and aromatic C=C skeletal vibration bands (Table 1). The UV spectra of IIIa-g are similar and do not contradict to the proposed structure (Table 1) since they have an absorption band characteristic for aromatic conjugated chromophores but lack the long-wavelength isoindole absorption band. The PMR spectral data are also in accord with the proposed structure (Table 2). Thus, the spectrum of IIIb recorded on spectrometer working at 500 MHz permitted to identify of all the aromatic protons. The signal of the olefinic proton ( $H_{\text{ol}}$ ) as triplet with a spin-spin coupling constant equal to that for the  $\text{CH}_2$  group doublet (2.3-2.7 Hz) is also clearly seen in this spectrum as well as in the PMR spectra for IIIa and IIIc taken at 100 MHz. We should note that diastereotopy for the methylene group protons is found in the PMR spectrum of IIIg taken in acetone- $d_6$  (see Experimental). These protons appear as two doublets with SSCC  $J = 22.0$  and 2.7 Hz. The phenomenon is explained by the noncoplanarity of the phenyl ring and double bond (see the discussion of the X-ray diffraction data for IIIc), which is apparently significant in solution in the case of bulky substituents. The splitting of these protons in the spectrum taken in  $\text{CDCl}_3$  is simplified and the two doublets of doublets degenerate into a doublet. The  $^{13}\text{C}$  NMR spectrum of IIIb has signals for all 26 carbon atoms (see Experimental). The appearance of three carbon atoms bonded to oxygen is confirmed and the atoms of the exocyclic double bond have been identified. The methyl and methylene group protons were found at high field. The use of programs to separate out the signals of  $\text{CH}_2$  group and quaternary carbon atoms from the signals of the methyl and methine group carbon atoms enabled us to assign all the signals in the spectrum of IIIb with complete proton decoupling.

The structure of IIIc was also proved by X-ray diffraction analysis (Fig. 1 and 2, Tables 3-5).\* This compound exists in the crystal as 2:1 solvate with acetone. The quinazoline fragment is planar. Deviation of the atoms from the mean square plane does not exceed 0.02 Å. Conjugation between the  $\pi$ -systems of the bicyclic system and the phenylene fragment is upset to a considerable extent due to twisting of the latter around the  $\text{C}_{(1)}-\text{C}_{(9)}$  bond by  $-71.8(5)^\circ$  (the  $\text{N}_{(1)}-\text{C}_{(1)}-\text{C}_{(9)}-\text{C}_{(10)}$  torsion angle) due to shortened intramolecular  $\text{H}_{(27\text{B})}-\text{C}_{(15)}$  contacts (2.82 Å) (the sum of the van der Waals radii is 2.87 Å [3]). This is also indicated by lengthening of the  $\text{C}_{(1)}-\text{C}_{(9)}$  bond to 1.494(5) Å relative to mean value (1.470 Å) [4]. These structural features of the compound studied indicate the possibility of atropoisomerism in series of derivative III. The  $\text{C}_{(15)}=\text{C}_{(16)}$  double bond is noncoplanar to the phenylene ring (the  $\text{C}_{(13)}-\text{C}_{(14)}-\text{C}_{(15)}-\text{C}_{(16)}$  torsion angle is  $-14.2(7)^\circ$ ), which probably results from short intramolecular contacts:  $\text{H}_{(27\text{B})}-\text{C}_{(15)}$  (2.82 Å) and  $\text{C}_{(13)}-\text{C}_{(19)}$  (3.18 Å) (the sum of the van der Waals radii is 3.42 Å),

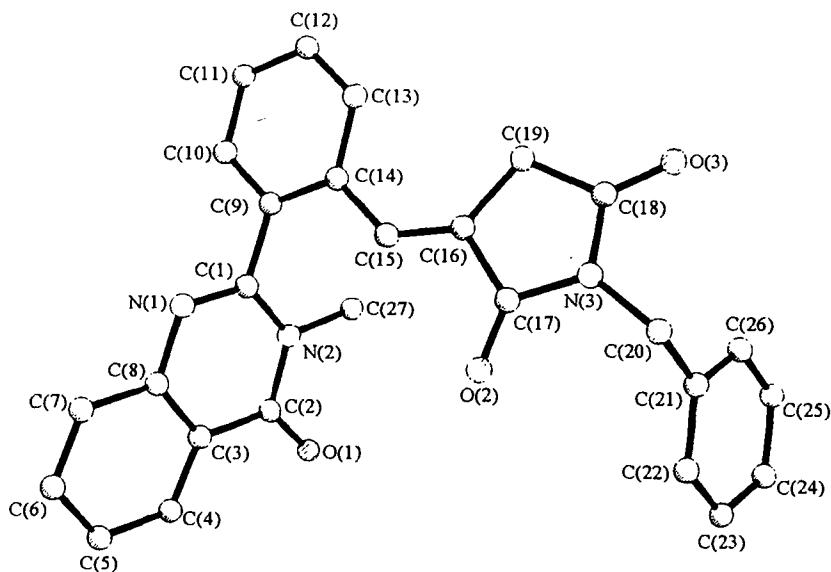


Fig. 1. General view of molecule of compound IIIc.

\*The numbering of the atoms in Fig. 1 does not correspond to the standard nomenclature.

TABLE 1. Characteristics of Synthesized Compounds

Compound	Empirical formula	Found, %			mp, °C	$R_f$	IR spectrum, $\nu, \text{cm}^{-1}$		UV spectrum		Yield, %
		C	H	N			Calculated, %		C=O	C=C	
IIIa	$\text{C}_{30}\text{H}_{15}\text{N}_3\text{O}_3$	69.12 69.56	4.43 4.38	12.41 12.17	225...226	0.28	1770, 1715, 1650	1610...1550	277	4.49	70
IIIb	$\text{C}_{36}\text{H}_{19}\text{N}_3\text{O}_3$	74.01 74.10	4.60 4.54	9.75 9.97	218...219	0.44	1775, 1715, 1670	1480...1440	1620...1560	286	4.46
IIIc	$\text{C}_{37}\text{H}_{21}\text{N}_3\text{O}_3$	74.55 74.47	4.91 4.86	9.54 9.65	193...194	0.47	1765, 1700, 1670	1515...1460	1610...1550	285	4.47
IIId	$\text{C}_{37}\text{H}_{21}\text{N}_3\text{O}_3$	74.32 74.47	4.93 4.86	9.70 9.65	233...234	0.46	1770, 1710, 1670	1615...1550	287	4.55	82
IIIE	$\text{C}_{36}\text{H}_{18}\text{N}_4\text{O}_5$	66.81 66.95	3.92 3.89	12.10 12.01	177...178	0.46	1760, 1710, 1670	1510...1450	1620...1570	299	4.63
IIIf	$\text{C}_{27}\text{H}_{21}\text{N}_3\text{O}_4$	71.92 71.83	4.63 4.69	9.39 9.31	224...225	0.43	1765, 1705, 1670	1505...1450	1610...1540	283	4.46
IIIG	$\text{C}_{30}\text{H}_{21}\text{N}_3\text{O}_3$	76.30 76.42	4.57 4.49	8.80 8.91	218...219	0.43	1770, 1705, 1640	1490...1440	1610...1570	285	4.52
											81

TABLE 2.  $^1\text{H}$  NMR Spectral Characteristics of Synthesized Compounds

Compound	Chemical shifts, $\delta$ , ppm						other protons	$^4J, \text{H}_{\text{ol}}, \text{CH}_2$	$^3J, 5\text{-H}, 6\text{-H}$	$^3J, o\text{-H}, m\text{-H}$
	$\text{N}-\text{CH}_3$ 3H, s	$\text{CH}_3$ , 2H, d	$\text{H}_{\text{ol}}$ , 1H, t	5-H, 1H, d	6-, 7-, 8-, 3'-, 4'-, 5'-, 6'-H	$\text{H}_{\text{atom}}$				
IIIa	3,28	3,70	7,32	8,26	7,46...7,97 (7H, m)	—	10,10 (1H, br. s, NH)	2,7	8,0	—
IIIb	3,22	3,91	7,25	8,23	7,59...7,95 (7H, m)	7,30 (2H, dd, o-H) 7,47 (2H, dd, m-H) 7,39 (1H, t, p-H)	—	2,3	8,0	8,5*
IIIc	3,30	3,58	7,47	8,37	7,20...7,90 (12H, m)* <sup>2</sup>	7,44...7,83 (8H, m)	4,71 (2H, s, <u><math>\text{CH}_2\text{Ph}</math></u> ) 2,36 (3H, s, $\text{CH}_3\text{C}_{\text{atom}}$ )	2,5	7,5	—
IIId	3,34	3,75	* <sup>3</sup>	8,36	7,45...7,90 (10H, m)* <sup>4</sup>	7,22 (4H, s) 8,32 (2H, d, o-H)	2,36 (3H, s, $\text{CH}_3\text{C}_{\text{atom}}$ )	2,5	7,5	—
IIIE	3,36	3,81	* <sup>3</sup>	8,37	7,32...8,05 (8H, m)	6,97 (2H, d, o-H)	—	2,7	8,0	9,0
IIIf	3,30	3,88	* <sup>3</sup>	8,27	7,15...8,06 (15H, m)* <sup>5</sup>	7,24 (2H, d, m-H)	3,80 (3H, s, $\text{OCH}_3$ )	2,5	8,0	8,7
IIIG	3,37	3,90	* <sup>3</sup>	8,35	—	—	—	2,5	8,0	—

\*  $^3J_{o\text{-H}} = 1.3$  and  $^3J_{m\text{-H}} = ^3J_{p\text{-H}} = 7.4$  Hz.\*<sup>2</sup> Signals for 5H<sub>atom</sub> of substituent R also included in the multiplet.\*<sup>3</sup> The signal is overlapped by the multiplet of 12H<sub>atom</sub> (IIId-f) of 15H<sub>atom</sub> (IIIfg).\*<sup>4</sup> The signals for H<sub>ol</sub> and 2m-H in R are also included in the multiplet.\*<sup>5</sup> The signals for H<sub>ol</sub> and 7H<sub>atom</sub> in R are also included in the multiplet.

and  $H_{(13)}-H_{(19)}$  (2.24 Å) (the sum of the van der Waals radii is 2.32 Å). These unfavorable interactions also lead to expansion of the  $C_{(14)}-C_{(15)}-C_{(16)}$  bond angle to 129.8(4)°. The five-membered ring has a flattened twist conformation. Atoms  $C_{(18)}$  and  $C_{(19)}$  are declined from the plane of  $C_{(16)}$ ,  $C_{(17)}$ , and  $N_{(3)}$  by -0.05 and 0.05 Å, respectively. The benzyl group is located almost perpendicularly to the mean plane of the five-membered ring (the  $C_{(17)}-N_{(3)}-C_{(20)}-C_{(21)}$  torsion angle is 83.9(5)°). The  $C_{(21)}\cdots C_{(26)}$  phenyl ring is twisted relative to the  $N_{(3)}-C_{(20)}$  bond by 67.6(5)° (the  $N_{(3)}-C_{(20)}-C_{(21)}-C_{(26)}$  torsion angle).

In the crystal, molecules of IIIc form channels containing the acetone solvate molecules (Fig. 2), which are disordered over two equally likely positions related to each other by a center of symmetry. The lack of shortened intermolecular contacts between the molecules of IIIc and acetone indicates that there are no clearly fixed positions of the solvate molecules and there are only areas of their predominant localization. This accounts for the relatively high values of the thermal parameters of the acetone atoms.

Formation of such unusual products as the result of the addition of maleimide derivatives to 6-methyl-5,6-dihydroisoindolo[2,1- $\alpha$ ]quinazolin-5-one system may be explained as follows. The reaction begins with the Diels – Alder cycloaddition leading to intermediate formation of adduct IV. The strain in intermediate IV leads to its rearrangement, occurring with dissociation of the bridge C–C and C–N and formation of two conjugated double bonds, one of which is exocyclic and the other is in the diazine ring.

The alternative formation of Michael adduct on the first stage does not create the preconditions for the rearrangement since there is no strain in such an adduct. Furthermore, in the case of formation of IIIa-g from the Michael adducts should give a mixture of their (*Z*)- and (*E*)- isomers, which is not observed in any case. The allylic coupling constant (2.3–2.5 Hz) clearly indicates a mutual *trans* arrangement of the  $CH_2$  group of the pyrrolidine ring and olefinic proton [5]. Precisely such structure should be obtained upon rearrangement of the Diels – Alder adduct of 6-methyl-5,6-dihydroisoindolo[2,1- $\alpha$ ]quinazolin-5-one and maleimide derivatives with bond breakage.

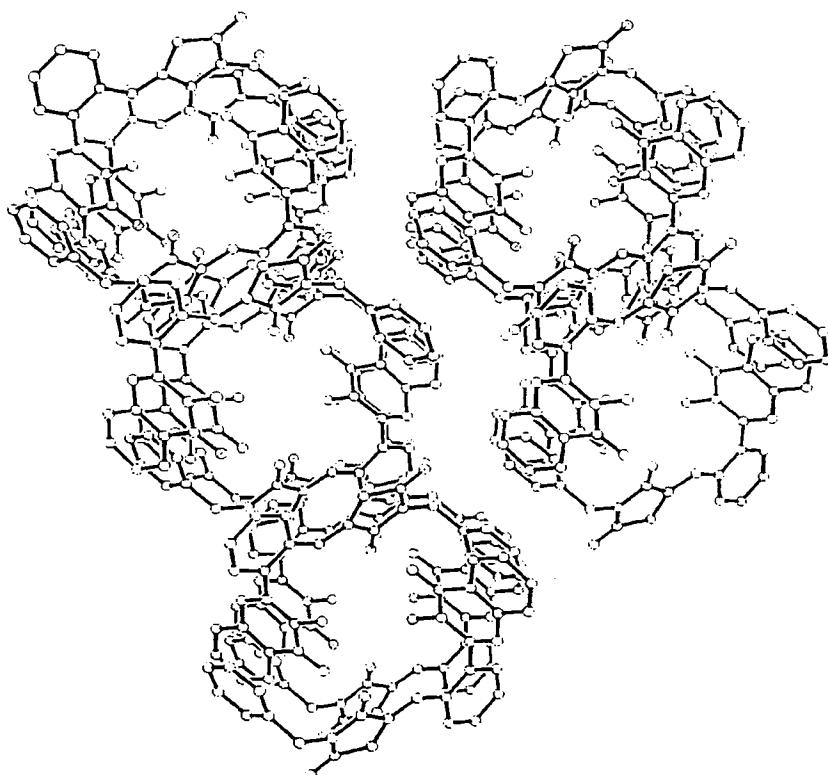


Fig. 2. Structure of molecules of compound IIIc in the crystal.

TABLE 3. Bond Lengths ( $d$ ) in the Structure of Compound IIIc

Bond	$d, \text{\AA}$	Bond	$d, \text{\AA}$
$\text{N}_{(1)}-\text{C}_{(1)}$	1,291(5)	$\text{C}_{(19)}-\text{C}_{(14)}$	1,395(5)
$\text{N}_{(1)}-\text{C}_{(8)}$	1,397(5)	$\text{C}_{(10)}-\text{C}_{(11)}$	1,377(6)
$\text{N}_{(2)}-\text{C}_{(1)}$	1,382(5)	$\text{C}_{(11)}-\text{C}_{(13)}$	1,373(6)
$\text{N}_{(2)}-\text{C}_{(2)}$	1,392(5)	$\text{C}_{(12)}-\text{C}_{(13)}$	1,380(6)
$\text{N}_{(2)}-\text{C}_{(27)}$	1,475(5)	$\text{C}_{(13)}-\text{C}_{(14)}$	1,403(5)
$\text{N}_{(3)}-\text{C}_{(18)}$	1,385(5)	$\text{C}_{(14)}-\text{C}_{(15)}$	1,454(5)
$\text{N}_{(3)}-\text{C}_{(17)}$	1,392(5)	$\text{C}_{(15)}-\text{C}_{(16)}$	1,334(5)
$\text{N}_{(3)}-\text{C}_{(20)}$	1,445(5)	$\text{C}_{(16)}-\text{C}_{(19)}$	1,482(6)
$\text{O}_{(1)}-\text{C}_{(2)}$	1,219(5)	$\text{C}_{(16)}-\text{C}_{(17)}$	1,495(5)
$\text{O}_{(2)}-\text{C}_{(17)}$	1,211(5)	$\text{C}_{(18)}-\text{C}_{(19)}$	1,504(6)
$\text{O}_{(3)}-\text{C}_{(18)}$	1,211(5)	$\text{C}_{(20)}-\text{C}_{(21)}$	1,500(6)
$\text{C}_{(1)}-\text{C}_{(9)}$	1,494(5)	$\text{C}_{(21)}-\text{C}_{(22)}$	1,369(7)
$\text{C}_{(2)}-\text{C}_{(3)}$	1,458(6)	$\text{C}_{(21)}-\text{C}_{(26)}$	1,386(6)
$\text{C}_{(3)}-\text{C}_{(8)}$	1,381(6)	$\text{C}_{(22)}-\text{C}_{(23)}$	1,387(7)
$\text{C}_{(3)}-\text{C}_{(4)}$	1,383(6)	$\text{C}_{(23)}-\text{C}_{(24)}$	1,378(8)
$\text{C}_{(4)}-\text{C}_{(5)}$	1,365(7)	$\text{C}_{(24)}-\text{C}_{(25)}$	1,368(7)
$\text{C}_{(5)}-\text{C}_{(6)}$	1,392(8)	$\text{C}_{(25)}-\text{C}_{(26)}$	1,373(7)
$\text{C}_{(6)}-\text{C}_{(7)}$	1,363(7)	$\text{O}_{(15)}-\text{C}_{(15)}$	1,22(1)
$\text{C}_{(7)}-\text{C}_{(8)}$	1,406(6)	$\text{C}_{(15)}-\text{C}_{(35)}$	1,50(1)
$\text{C}_{(9)}-\text{C}_{(10)}$	1,393(5)	$\text{C}_{(15)}-\text{C}_{(25)}$	1,50(1)

TABLE 4. Bond Angles ( $\phi$ ) in the Structure of Compound IIIc

Angle	$\phi, \text{deg.}$	Angle	$\phi, \text{deg.}$
$\text{C}_{(1)}-\text{N}_{(1)}-\text{C}_{(8)}$	117,1(4)	$\text{C}_{(12)}-\text{C}_{(13)}-\text{C}_{(14)}$	120,9(4)
$\text{C}_{(1)}-\text{N}_{(2)}-\text{C}_{(27)}$	120,9(4)	$\text{C}_{(9)}-\text{C}_{(14)}-\text{C}_{(13)}$	117,7(4)
$\text{C}_{(1)}-\text{N}_{(2)}-\text{C}_{(2)}$	121,6(3)	$\text{C}_{(9)}-\text{C}_{(14)}-\text{C}_{(15)}$	119,8(4)
$\text{C}_{(2)}-\text{N}_{(2)}-\text{C}_{(27)}$	117,4(4)	$\text{C}_{(13)}-\text{C}_{(14)}-\text{C}_{(15)}$	122,5(4)
$\text{C}_{(18)}-\text{N}_{(3)}-\text{C}_{(17)}$	112,6(3)	$\text{C}_{(16)}-\text{C}_{(15)}-\text{C}_{(14)}$	129,8(4)
$\text{C}_{(18)}-\text{N}_{(3)}-\text{C}_{(20)}$	123,2(3)	$\text{C}_{(15)}-\text{C}_{(16)}-\text{C}_{(19)}$	132,5(4)
$\text{C}_{(17)}-\text{N}_{(3)}-\text{C}_{(20)}$	124,2(4)	$\text{C}_{(15)}-\text{C}_{(16)}-\text{C}_{(17)}$	120,3(4)
$\text{N}_{(1)}-\text{C}_{(1)}-\text{N}_{(2)}$	124,6(4)	$\text{C}_{(19)}-\text{C}_{(16)}-\text{C}_{(17)}$	107,2(3)
$\text{N}_{(1)}-\text{C}_{(1)}-\text{C}_{(9)}$	116,9(4)	$\text{O}_{(2)}-\text{C}_{(17)}-\text{N}_{(3)}$	123,8(4)
$\text{N}_{(2)}-\text{C}_{(1)}-\text{C}_{(9)}$	118,6(3)	$\text{O}_{(2)}-\text{C}_{(17)}-\text{C}_{(16)}$	129,3(4)
$\text{O}_{(1)}-\text{C}_{(2)}-\text{N}_{(2)}$	120,6(4)	$\text{N}_{(3)}-\text{C}_{(17)}-\text{C}_{(16)}$	106,9(4)
$\text{O}_{(1)}-\text{C}_{(2)}-\text{C}_{(3)}$	124,7(4)	$\text{O}_{(3)}-\text{C}_{(18)}-\text{N}_{(3)}$	124,4(4)
$\text{N}_{(2)}-\text{C}_{(2)}-\text{C}_{(3)}$	114,7(4)	$\text{O}_{(3)}-\text{C}_{(18)}-\text{C}_{(19)}$	127,6(4)
$\text{C}_{(8)}-\text{C}_{(3)}-\text{C}_{(4)}$	120,4(4)	$\text{N}_{(3)}-\text{C}_{(18)}-\text{C}_{(19)}$	108,1(3)
$\text{C}_{(8)}-\text{C}_{(3)}-\text{C}_{(2)}$	119,1(4)	$\text{C}_{(16)}-\text{C}_{(19)}-\text{C}_{(18)}$	104,7(3)
$\text{C}_{(4)}-\text{C}_{(3)}-\text{C}_{(2)}$	120,5(5)	$\text{N}_{(3)}-\text{C}_{(20)}-\text{C}_{(21)}$	114,3(4)
$\text{C}_{(5)}-\text{C}_{(4)}-\text{C}_{(3)}$	120,2(5)	$\text{C}_{(22)}-\text{C}_{(21)}-\text{C}_{(26)}$	117,7(5)
$\text{C}_{(4)}-\text{C}_{(5)}-\text{C}_{(6)}$	120,0(5)	$\text{C}_{(22)}-\text{C}_{(21)}-\text{C}_{(20)}$	121,4(4)
$\text{C}_{(7)}-\text{C}_{(6)}-\text{C}_{(5)}$	120,4(5)	$\text{C}_{(26)}-\text{C}_{(21)}-\text{C}_{(20)}$	120,8(4)
$\text{C}_{(6)}-\text{C}_{(7)}-\text{C}_{(8)}$	119,8(5)	$\text{C}_{(21)}-\text{C}_{(22)}-\text{C}_{(23)}$	121,3(5)
$\text{C}_{(3)}-\text{C}_{(8)}-\text{N}_{(1)}$	122,7(4)	$\text{C}_{(24)}-\text{C}_{(23)}-\text{C}_{(22)}$	120,3(5)
$\text{C}_{(3)}-\text{C}_{(8)}-\text{C}_{(7)}$	119,1(4)	$\text{C}_{(25)}-\text{C}_{(24)}-\text{C}_{(23)}$	118,6(5)
$\text{N}_{(1)}-\text{C}_{(8)}-\text{C}_{(7)}$	118,1(4)	$\text{C}_{(24)}-\text{C}_{(25)}-\text{C}_{(26)}$	121,0(5)
$\text{C}_{(10)}-\text{C}_{(9)}-\text{C}_{(14)}$	120,8(4)	$\text{C}_{(25)}-\text{C}_{(26)}-\text{C}_{(21)}$	121,1(5)
$\text{C}_{(10)}-\text{C}_{(9)}-\text{C}_{(1)}$	117,9(4)	$\text{O}_{(15)}-\text{C}_{(15)}-\text{C}_{(35)}$	117,0(4)
$\text{C}_{(14)}-\text{C}_{(9)}-\text{C}_{(1)}$	121,2(3)	$\text{O}_{(15)}-\text{C}_{(15)}-\text{C}_{(25)}$	110,0(3)
$\text{C}_{(11)}-\text{C}_{(10)}-\text{C}_{(9)}$	120,2(4)	$\text{C}_{(35)}-\text{C}_{(15)}-\text{C}_{(25)}$	132,0(5)
$\text{C}_{(12)}-\text{C}_{(11)}-\text{C}_{(10)}$	119,9(4)	$\text{C}_{(11)}-\text{C}_{(12)}-\text{C}_{(13)}$	120,6(4)

TABLE 5. Coordinates ( $\times 10^4$ ) and Equivalent Isotropic Temperature Factors ( $\text{\AA}^2 \times 10^3$ ) of the Non-Hydrogen Atoms in the Structure of Compound IIIc

Atom	<i>x</i>	<i>y</i>	<i>z</i>	<i>U</i> <sub>eq</sub>
N <sub>(1)</sub>	4223(4)	6007(4)	8178(2)	51(1)
N <sub>(2)</sub>	5141(4)	5186(4)	6951(2)	46(1)
N <sub>(3)</sub>	10(4)	4489(4)	2922(2)	47(1)
O <sub>(1)</sub>	7331(4)	6663(4)	6717(3)	72(1)
O <sub>(2)</sub>	1980(4)	6382(3)	4260(2)	54(1)
O <sub>(3)</sub>	-1831(4)	2211(4)	1906(2)	83(1)
C <sub>(1)</sub>	4095(5)	5004(4)	7453(3)	43(1)
C <sub>(2)</sub>	6447(6)	6512(5)	7191(3)	49(1)
C <sub>(3)</sub>	6641(5)	7646(4)	8033(3)	47(1)
C <sub>(4)</sub>	7947(6)	8975(5)	8399(4)	71(2)
C <sub>(5)</sub>	8134(7)	10022(6)	9199(4)	83(2)
C <sub>(6)</sub>	7000(7)	9762(5)	9640(4)	77(2)
C <sub>(7)</sub>	5711(6)	8453(5)	9292(3)	68(2)
C <sub>(8)</sub>	5524(6)	7360(4)	8480(3)	49(1)
C <sub>(9)</sub>	2711(5)	3553(4)	7143(3)	42(1)
C <sub>(10)</sub>	2630(6)	2643(5)	7843(3)	58(1)
C <sub>(11)</sub>	1354(6)	1318(5)	7609(4)	66(2)
C <sub>(12)</sub>	153(6)	901(5)	6690(4)	61(1)
C <sub>(13)</sub>	208(5)	1803(4)	5997(3)	52(1)
C <sub>(14)</sub>	1496(5)	3155(4)	6209(3)	44(1)
C <sub>(15)</sub>	1581(5)	4149(4)	5510(3)	44(1)
C <sub>(16)</sub>	700(5)	3906(4)	4522(3)	45(1)
C <sub>(17)</sub>	1035(5)	5103(5)	3944(3)	45(1)
C <sub>(18)</sub>	-930(6)	2999(5)	2751(3)	55(1)
C <sub>(19)</sub>	-626(5)	2576(4)	3783(3)	54(1)
C <sub>(20)</sub>	-45(6)	5294(5)	2111(3)	59(1)
C <sub>(21)</sub>	1076(6)	5394(5)	1568(3)	53(1)
C <sub>(22)</sub>	2271(7)	6714(6)	1665(4)	72(2)
C <sub>(23)</sub>	3273(7)	6811(6)	1132(4)	84(2)
C <sub>(24)</sub>	3100(7)	5564(7)	504(4)	74(2)
C <sub>(25)</sub>	1922(7)	4240(6)	419(3)	69(2)
C <sub>(26)</sub>	923(6)	4149(5)	939(3)	62(1)
C <sub>(27)</sub>	4944(6)	3953(5)	6167(3)	62(1)
O <sub>(15)</sub>	3648(28)	10195(20)	5610(21)	233(10)
C <sub>(15)</sub>	4632(32)	10056(21)	5327(20)	164(15)
C <sub>(25)</sub>	4556(32)	10478(26)	4301(23)	187(14)
C <sub>(35)</sub>	5420(85)	9247(70)	5910(47)	680(102)

## EXPERIMENTAL

The <sup>1</sup>H NMR spectra were recorded on a Bruker-500 spectrometer for IIIb and on a Bruker-100 spectrometer in CDCl<sub>3</sub> for IIIc-e,g and in acetone-d<sub>6</sub> for IIIa,f,g relative to TMS. The <sup>13</sup>C NMR spectrum of IIIb in DMSO-d<sub>6</sub> was taken on a Bruker-500 spectrometer at 125.75 MHz. The IR spectra were recorded on a Pye-Unicam SP3-300 spectrometer for KBr pellets. The UV spectra were obtained on a Specord UV-VIS spectrometer in 1,2-dichloroethane. The melting points were found on a Boetius block. The mass spectrum of IIIb was recorded on a Finnigan MAT-8200 mass spectrometer. The reaction course and purity of the products were monitored by thin-layer chromatography on Silufol UV-254 plates using 2:1 benzene-acetone as the eluent. The characteristics of the products are given in Tables 1 and 2.

**6-Methyl-5,6-dihydroisoindolo[2,1-*a*]quinazolin-5-one** was obtained according to Babichev [6].

**X-ray Structural Analysis of the Solvate of Compound IIIc, 0.5C<sub>3</sub>H<sub>6</sub>O(C<sub>27</sub>H<sub>21</sub>N<sub>3</sub>O<sub>3</sub>).** The unit cell parameters of triclinic crystals of the solvate of dihydroquinazoline IIIc at 20°C: *a* = 10.149(3), *b* = 10.158(3), *c* = 13.699(4) Å,  $\alpha$  = 95.69(3)°,  $\beta$  = 106.13(2)°,  $\gamma$  = 113.29(2)°, *V* = 1210.7(6) Å<sup>3</sup>, *d*<sub>calc</sub> = 1.283 g/cm<sup>3</sup>, space group

P1,  $Z = 2$ . The unit cell parameters and intensities of 2912 independent reflections ( $R_{\text{int}} = 0.031$ ) were measured on a Siemens P3/PC automatic diffractometer using  $\lambda\text{MoK}_{\alpha}$  radiation, graphite monochromator, and  $\theta/2\theta$  scanning,  $2\theta_{\text{max}} = 50^\circ$ .

The structure was solved by the direct method using the SHELXTL PLUS program package [7]. The positions of the hydrogen atoms were calculated geometrically and refined by the "horseback rider" model with fixed  $U_{\text{iso}} = nU_{\text{eq}}$  of the non-hydrogen atom attached to the given hydrogen atom ( $n = 1.5$  for methyl groups and 1.2 for the other hydrogen atoms). The acetone solvate molecule was refined assuming equal C–C (1.50(1) Å) and C–O bond lengths (1.21 Å). Anisotropic refinement for  $F^2$  by the full-matrix least-squares method using 2514 reflections for the non-hydrogen atoms was carried out to  $wR_2 = 0.168$  ( $R_1 = 0.072$  using 2302 reflections with  $F > 4\sigma(F)$ ,  $S = 1.21$ ). The coordinates of the non-hydrogen atoms are given in Table 5.

**General Method for the Preparation of 2-{2-[(E)-1-R-2,5-Dioxopyrrolidinidenemethyl]phenyl}-3-methyl-4-oxo-3,4-dihydroquinazolines (IIIa-g).** A mixture of corresponding maleimide II (2 mmol) and compound I (2 mmol) was refluxed in 15–20 ml of 2-propanol for 1 h (for IIe) and for 2 h (for IIIa-d,f,g). The reaction mixture was cooled. The precipitate of compound III was filtered off and recrystallized from a mixture of acetone and 2-propanol.

Compound IIIg,  $^1\text{H}$  NMR spectrum in acetone- $d_6$ : 3.35 (3H, s, N–CH<sub>3</sub>), 3.97 (1H, dd,  $^2J = 22.0$  and  $^4J = 2.7$  Hz, CH<sub>a</sub>H<sub>b</sub>), 4.26 (1H, dd,  $^4J = 2.7$  Hz, CH<sub>a</sub>CH<sub>b</sub>), 7.35–8.15 (15H, m), 8.26 ppm (1H, d,  $^3J = 7.8$  Hz, 5-H).

Compound IIIb,  $^{13}\text{C}$  NMR spectrum: 32.904 (N–CH<sub>3</sub>), 33.948 (CH<sub>2</sub>), 95.367, 120.249, 126.163, 126.928, 126.965, 127.255, 127.339, 127.716, 128.036, 128.208, 128.632, 128.694, 128.931, 129.932, 130.213, 131.469, 132.327, 134.578 (C=C), 136.436, 146.764 (C<sub>arom</sub>–N), 154.548 (C<sub>(2)</sub>), 161.209, 169.416 and 173.210 ppm (C=O). Mass spectrum,  $m/z$  (I, %): 421 (100), 406 (3), 392 (19), 330 (4), 301 (7), 274 (85), 259 (10), 248 (31), 233 (9), 229 (4), 216 (4), 205 (4), 151 (6), 140 (4), 137 (6), 124 (4), 77 (4).

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