

Electron Deficient Heteroaromatic Ammonioamidates. Part 24.¹ *N*-(Quinazolin-3-*io*)amidates. Part 11.² The Photochemistry of *N*-(6,7-Methylenedioxyquinazolin-3-*io*)amidates in Acetone

Gizella Barta-Szalai, József Fetter, and Károly Lempert *

Department of Organic Chemistry, Technical University Budapest, H-1521 Budapest, Gellért tér 4, Hungary

Jørgen Møller

Department of Chemistry, Odense University, Campusvej 55, DK-5230 Odense, Denmark

László Párkányi

Central Research Institute for Chemistry of the Hungarian Academy of Sciences, H-1525 Budapest, Hungary

N-(Quinazolin-3-*io*)amidates (1) may exist, depending on the nature of the groups R, R¹, and R², and in the absence of nucleophiles, as equilibrium mixtures of the monomeric (1) and dimeric (3) forms. For the first time evidence has been found for the generation of photoproducts of the dimeric forms (3) *via* irradiation of the quinazolinioamidates (1a–c) in acetone in which substantial amounts of the dimers (3a–c) are present. Thus, the quinazolinioamidates (1) are the only heteroaromatic ammonioamidates which are known not only to exist in three forms, *viz.* the monomer, the adducts (2), and the dimers (3), but also to furnish characteristic photoproducts of all three forms.

Of the electron deficient heteroaromatic *N*-ammonioamidates studied so far the *N*-(quinazolin-3-*io*)amidates (1) †‡ are the only ones which (to a greater or lesser degree, depending on the nature of the ligands R, R¹, and R², as well as the Nu group) are able to form adducts (2) with nucleophiles HNu and, in the absence of such nucleophiles, dimers (3),^{4–8} As a consequence, the photochemistry of compounds (1) is more complicated than that of other ammonioamidates. In the presence of nucleophiles (*e.g.* alcohols,⁶ toluene- α -thiol,⁷ amines,⁸ and acetamide⁸) two series of characteristic photoproducts, originating from forms (1) and (2), respectively, are formed.

Here we report the photochemistry of a series of *N*-(quinazolin-3-*io*)amidates (1a–h) § in acetone; this solvent, in the absence of acidic and basic catalysts,⁹ is unable to form type (2) adducts, *viz.* (13), with the amidates. Compounds (1a–h) were irradiated in acetone under argon with a high-pressure mercury immersion lamp (HPK-125) through Pyrex. Chromatographic work-up of the resulting mixtures furnished the photoproducts (4)–(13) (Table 1). ¶ The majority of the photoproducts had been obtained previously on irradiation of compounds (1a–h) in the presence of nucleophiles as shown in Table 2. On the other hand, compounds (9h), (11a–c), and (12e) are novel and such products have not been obtained before on irradiation of heteroaromatic ammonioamidates. Compounds (7d) and (7h) are also new. (For proof of the structures of the new compounds, see below.)

† For a review on the chemistry of *N*-(quinazolin-3-*io*)amidates and related compounds with special emphasis on their photochemistry (see ref. 3).

‡ Compounds of this type are correctly named as 'amidides'; they are so named in the Experimental section.

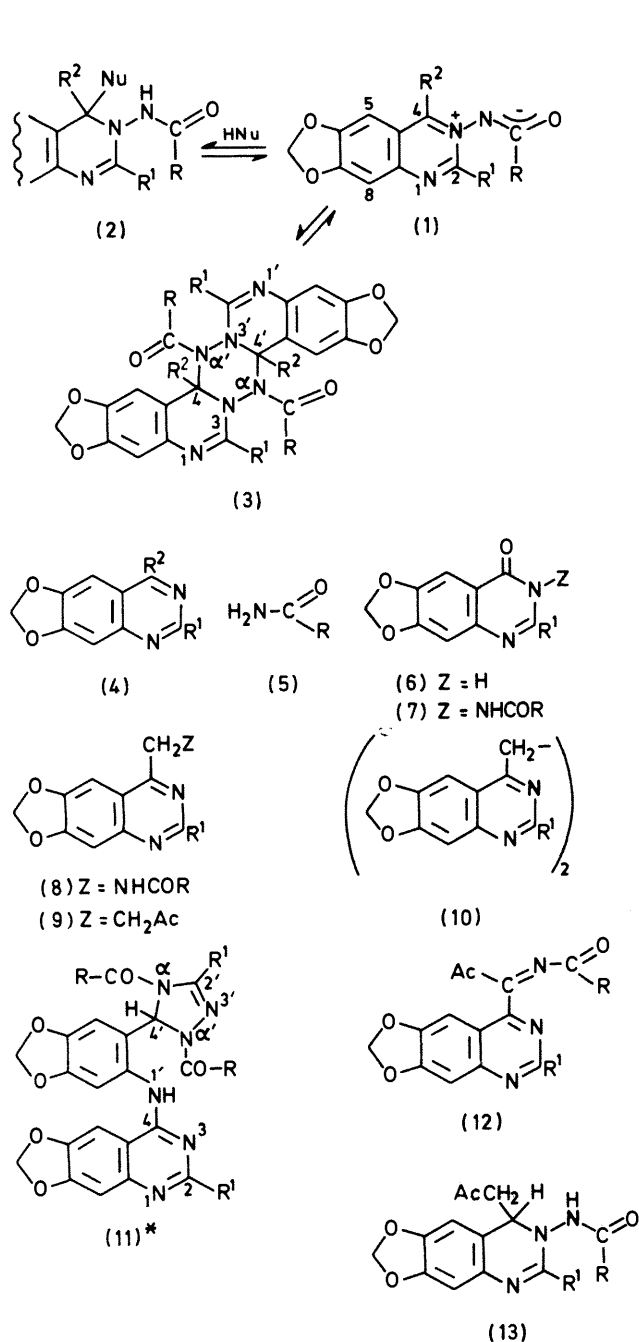
§ Compounds (1a–c) exist in the crystalline state as the pure dimer (3a) and as mixtures of the monomers (1b), (1c) and the dimers (3b), (3c), respectively. However, in CDCl₃ solution equilibria between the monomeric and dimeric forms are rapidly established.⁴ This has been shown to be the case for the amidate (1b), and may be assumed for the amidates (1a) and (1c) in solvent acetone as well. It is, thus, immaterial whether monomers or dimers are actually introduced as the starting materials in the (a)–(c) series.

¶ Although irradiation was continued until no unchanged starting compounds were detected in the reaction mixture by t.l.c., type (13) compounds may be by-products formed during work-up from unchanged starting material.^{9,10}

Compounds (6) and (7) are clearly photo-oxidation products, their formation being the result of the presence of traces of oxygen and, possibly, water in the system (*cf.* ref. 6). Some type (7) products are also formed *via* autooxidation of quinazolinioamidates (1) in the dark.¹⁰ Compounds of type (4), (8), and (10) have been shown to be derived from the monomeric quinazolinioamidates.^{6–8} The amides (5) are the co-products of the quinazolines (4).

While compounds (9h) and (12e) may be considered to be photoproducts of the monomeric amidates (1h) and (1e), respectively, incorporating an acetyl group originating from the solvent acetone [see Scheme 1 for a possible route to (9h)] compounds (11) are assumed to be photoproducts of the dimeric forms (3), although other routes for the formation of these compounds are not ruled out by the available experimental facts. This assumption is based on the observation that the products (11) are obtained only under conditions in which the quinazolinioamidates (1) form the dimers (3) rather than adducts of type (2) with the solvent; also, they have never been obtained in the presence of nucleophiles, when the starting compounds exist as equilibrium mixtures (1) \rightleftharpoons (2) rather than as mixtures (1) \rightleftharpoons (3). Further support comes from the observation that the photoproducts (11) were obtained only from those compounds (1a–c) which [in contrast to compounds (1e–g)] are known to exist partly as the dimers (3a–c) in chloroform solution. It has now been shown that compound (1d) does *not* form a dimer in chloroform. Moreover, compound (1b) exists in acetone in the form of a *ca.* 4 : 1 dimer–monomer equilibrium. Because of the very low solubility of compounds (1d) and (1e) in acetone, the position of the dimer–monomer equilibrium could not be determined but, for (1e), appears to be strongly shifted towards the monomeric form.

It was therefore considered important to establish conclusively that the second form of the quinazolinioamidates (1), present in solvents which do not form adducts of type (2), is indeed the dimer (3). Although the dimeric structures had been rigorously established for the crystalline state (*X*-ray diffraction, *i.r.* spectra),^{4,5} the ¹H n.m.r. evidence which was presented as proof for the dimeric structures in solution⁴ is actually ambiguous: the ¹H n.m.r. spectra are also consistent with the presence of the tautomeric forms (14) of the monomers (1). The final evidence for the dimeric form (3) rather than the tautomeric structures (14) came from a ¹³C n.m.r.

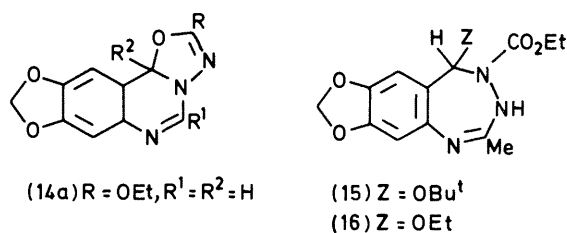


* Numbering shows origin from compounds (3)

| | R | (1)–(13) R ¹ | R ² | Ref. |
|----|---------------------|----------------------------|----------------|------|
| a; | OEt | H | H | 4 |
| b; | OEt | Me | H | 4 |
| c; | OCH ₂ PH | Me | H | 4 |
| d; | Me | Me | H | 1 |
| e; | Ph | Me | H | 4 |
| f; | OEt | H | Me | 4 |
| g; | OEt | Me | Me | 4 |
| h; | Ph | Me | Me | 6 |

study: the low-field signal at 156.2 p.p.m. in the CCl₄ spectrum of compound (1a) (which is known⁴ to exist in this solvent in a single form) is characteristic of an N-C(O)OEt group and therefore definitely rules out the tautomeric structure (14a).

Scheme 1. ^a Cf. ref. 6



In spite of considerable effort we were unable to isolate any intermediates from the photoisomerizations (3) \rightarrow (11).

For comparison, some irradiations were carried out in other non-nucleophilic solvents as well as in *t*-butyl alcohol (Table 3), which does not form type (2) adducts either because of the bulk of the *t*-butoxy group.⁴ The benzotriazine (15) [whose ethoxy analogue (1b) had been obtained on irradiation of compound (1b) in ethanol⁶] was the only photoproduct which had not occurred from the irradiations carried out in acetone. It had been shown that compound (16) was derived from the monomeric amidate (1b)⁶ and, by analogy, this is thought to be the case for compound (15) as well.

Elucidation of the Structures of the New Photoproducts.—The structure of compound (7d) was deduced from its i.r., ¹H n.m.r., and mass spectra while (7h) [= (7e)] was identified by comparison with an authentic sample. The structure of compound (9h) was deduced from its mass and ¹H n.m.r. spectra and that of compound (11b) was elucidated by an X-ray diffraction study. The structures of compounds (11a) and (11c) were tentatively obtained on the basis of the close similarity of the ¹H n.m.r. spectra of (11a–c). The mass spectra of these three compounds exhibit the correctly corresponding molecular ions.

The structure of compound (12e) was tentatively assigned on the basis of its mass spectrum and substantiated by structure-proving synthesis (see below). The structure of compound (15) follows from the close similarity of its ¹H n.m.r. spectrum to that of the analogous compound (16).

Table 1. Irradiation products of compounds (1a–h) in acetone ^{a,b}

| Starting compound (mmol) | Acetone [cm ³ /mmol (1)] | Irradiation time (h) | Isolated products and yields (%) | | | | | | | | | |
|--------------------------|-------------------------------------|----------------------|----------------------------------|-------------|------------|--------------------|-----------------|----------------|------|----------------------|------------|----------------------------|
| | | | (4) | (5) | (6) | (7) | (8) | (9) | (10) | (11) | (12) | (13) |
| (1a) 5.75 | 28 | 20 | 10 (C) | | | 10 (B) | | | | 12 (C) | | 14 (B) |
| 11.5 | 43 | 50 | 17 (C) | <i>c</i> | | 15 (B) | | | | 5.5 (C) | | <i>d</i> |
| (1b) 5.45 | 27 | 20 ^e | 12–17 (C) | | 1.5 (C) | | | | | 10–17 (C) | | 18 (B + C) ^d |
| (1c) 4.5 | 34 | 20 | 8.5 (C) | | | | | | | 12 (C) | | 19 (B) |
| 8.9 | 34 | 25 | 44 (C) | 22 (C) | | 5.5 (C) | | | | 8.5 (C) | | |
| (1d) 4.1 | 61 | 30 | 39 (B) | <i>c</i> | | 19 (B + C) | | | | | | |
| (1e) 4.9 | 51 | 60 | 20 (C) | 15 (C) | | | | | | | 8.5 (C) | |
| (1f) 2.5 | 60 | 2 | | | | 7.5 (B + C) | 11.5 (B + C) | | | 32.5 (A + B C) | | |
| (1g) 1.7 | 87 | 6 | 17 (C) | | | | 24 (C) | | | 16 (A + B) | | |
| (1h) 2.5 | 40 | 12 ^f | 39 (B + C) | 4.5 (C) | | | | 4 (B + C) | | 32 (A) | | |
| 3.4 | 44 | 12 | 27 (B + C) | 28.5 (C) | | | | 7.5 (B + C) | | 33 (A) | | |
| 9.4 | 32 | 25 | 40.5 (C) | 46 (C) | | trace ^e | | 1.5 (C) | | 38 (A) | | |

^a Compounds (1a–c) exist as both monomers and dimers (3a–c) in acetone (see Discussion section). ^b For the reaction conditions and the details of the work-up procedure, fractions A–C, see Experimental section. ^c Detected by t.l.c. ^d Several products of unknown structures were also obtained. ^e Several runs. ^f Irradiation through quartz.

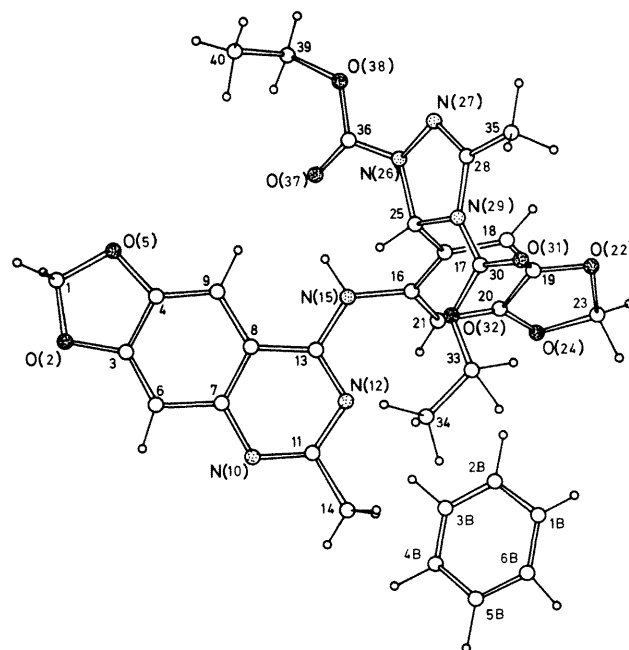
Table 2. Some products of irradiation of compounds (1a–h) in various solvents ^{a,b}

| Solvent | (4) | (5) | (7) | (8) | (10) |
|---|--|-------------------------------|--------------------|-------------------|------------------|
| Ethanol | b–c ⁶ d ¹ e–h ⁶ | b, c, e, g, h ⁶ | a–c ^{6,c} | f, g ⁶ | f–h ⁶ |
| Benzyl alcohol | b ⁶ | | | | |
| Toluene- α -thiol- chloroform | e–h ⁷ | a, b, e–h ⁷ | a ⁷ | g ⁷ | |
| Butylamine | g–h ⁸ | g–h ⁸ | | | |

^a Photoproducts originating from the type (2) adducts and those whose formation requires participation of the nucleophile present are not listed. ^b Compounds (1a–c) exist as both monomers and dimers (3a–c) in acetone (see Discussion section). ^c Compounds (7a) and (7b) were also obtained in aqueous dioxane and aqueous ethanol, respectively. ⁶

Structure Determination of Compound (11b) by X-Ray Diffraction.—The structure of compound (11b) (Figure) has been derived from three-dimensional X-ray diffractometry data. The sample used for the structure determination contained one molecule of benzene of crystallization for each molecule of compound (11b). The atomic co-ordinates, bond lengths, and bond angles with their e.s.d.s are listed in Tables 4–6. Tables of structure factors are available as a Supplementary Publication * (SUP No. 23612, 24 pages).

Structure-proving Synthesis of Compound (12e).—The synthesis of compound (12e) is shown in Scheme 2. The acetone ad-

**Figure.** Perspective view of compound (11b) with crystallographic numbering

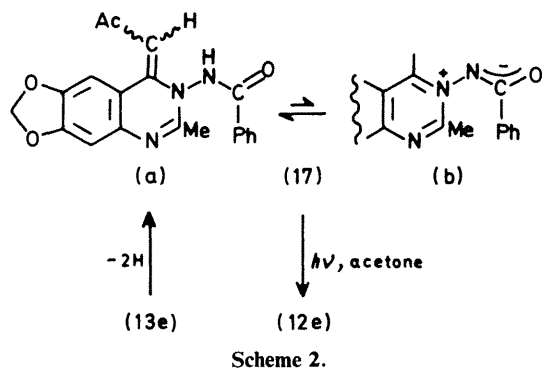
duct (13e) ⁹ of compound (1e) was dehydrogenated to (17) with alkaline potassium hexacyanoferrate(III); attempted dehydrogenation by irradiation in acetone under oxygen furnished compound (7e) rather than the desired (17). As shown by its ¹H n.m.r. spectrum, compound (17) does exist in deuterio-

* For details of the Supplementary Publications Scheme, see Instructions to Authors (1983), *J. Chem. Soc., Perkin Trans. I*, 1983, Issue 1.

Table 3. Irradiation of some compounds of type (1) in various solvents ^{a,b}

| | Starting compound (mmol) | Solvent [cm ³ /mmol (1)] | Irradiation time (h) | Isolated products and yields (%) | | | | | | |
|------|--------------------------|-------------------------------------|----------------------|----------------------------------|--------------|----------|---------|----------|---------|--------------|
| | | | | (4) | (5) | (6) | (7) | (10) | (11) | (15) |
| (1a) | 5.75 | CH ₂ Cl ₂ | 26 | 80 | 23 (B) | | | 6.5 (B) | | |
| | 11.5 | CH ₂ Cl ₂ | 35 | 100 | 25 (C) | 9.5 (B) | | 14.5 (B) | | |
| (1b) | 5.45 | CH ₂ Cl ₂ | 37 | 70 | 31.5 (B + C) | <i>c</i> | 3 (C) | 5 (C) | | 2 (C) |
| | | CH ₂ Cl ₂ | 28 | 30 | 9 (C) | <i>c</i> | | | | 15.5 (B + C) |
| (1b) | 1.8 | MeCN | 83 | 15 | 32.5 (C) | <i>c</i> | 4.5 (C) | | | 22 (C) |
| (1b) | 5.45 | Bu ^o OH | 59 | 15 | 5 (C) | <i>c</i> | | | | 14 (C) |
| (1e) | 1.65 | CH ₂ Cl ₂ | 12 | 15 | 35.5 (B) | 32 (C) | | | | 10 (C) |
| (1h) | 4.65 | CH ₂ Cl ₂ | 13 | 15 | 32 (B) | 33.5 (B) | | | | |
| (1h) | 1.95 | HCO ₂ Et | 76 | 2 | 70 (C) | 67 (C) | | | 6.0 (A) | |

^a Compounds (1a–c) exist as both monomers and dimers (3a–c) in acetone (see Discussion section). ^b For the reaction conditions and the details of the work-up procedure and fractions A–C, see Experimental section. ^c Detected by t.l.c.



chloroform solution exclusively in the form of the methylene base (17a). Irradiation of compound (17) in acetone furnished compound (12e) by N → C migration of the benzoylamino group and concomitant dehydrogenation. Similar migrations of ethoxycarbonylamino groups (without dehydrogenation) have been observed before^{6,11} but the photorearrangement (17) → (12e) is the first instance of the migration of a benzoylamino and, more generally, of a simple acylamino group. It is thus noteworthy that compound (8h), in spite of all our efforts, was never detected in the irradiation mixtures of compound (1h); instead compound (9h) was isolated. As shown by a separate experiment, (9h) is *not* a secondary phototransformation product of compound (4h). The probable route for the formation of compound (9h) is shown in Scheme 1.

Experimental

¹H and ¹³C n.m.r. spectra were obtained with a JEOL FX-100 spectrometer, unless otherwise stated, for solutions in CDCl₃ at room temperature. [²H₆]Acetone–[²H₆]benzene (contained in a separate tube placed coaxially in the tube containing the solution of the compound studied) was used as the D-lock when CCl₄ was used as the solvent. Mass spectra were obtained on a Varian MAT 311A instrument (Grant from the Danish Natural Science Research Council) by electron impact (70 eV) using the direct insertion system.

¹³C N.M.R. Spectrum of N-(6,7-Methylenedioxyquinazolin-3-yl)ethoxyformamide (1a) in CCl₄. Proof that It exists exclusively as the Dimer (3a).—The ¹³C n.m.r. spectrum of (1a) at 50 °C was as follows: δ 16.1 (q) * and 64.5 (t) (OEt), 75.1

(d, N–CH–N), 102.6 (t, OCH₂O), 107.6 (d), 108.8 (d), 115.6 (s), 136.2 (s), 146.3 (s), 147.7 (s), and 150.5 (s) (ArC and N=C–N), and 156.2 p.p.m. [s, N–C(O)OEt].

¹H N.M.R. Spectra of Compounds (1b), (1d), and (1e).—(a) N-(2-Methyl-6,7-methylenedioxyquinazolin-3-yl)ethoxyformamide (1b), in [²H₆]acetone, gave two series of signals, corresponding to the monomer (1b) and the dimer (3b), respectively; δ (1.32 (t) and 4.05 (q) (CO₂Et), 2.86 (s, 2-Me), 6.36 (s, OCH₂O), 7.31 (s) and 7.44 (s) (5- and 8-H), and 9.86 (s, 4-H); δ 1.31 (t) and 4.32 (q) (CO₂Et), 1.68 (s, 2-Me), 6.00 (s, OCH₂O), and 6.58 (s), 6.88 (s) and 7.00 (s) (4-, 5-, and 8-H) p.p.m., respectively. The intensity ratios of the corresponding signals were *ca.* 1 : 4.

(b) N-(2-Methyl-6,7-methylenedioxyquinazolin-3-yl)acetamide (1d), in CDCl₃ (purified by treatment with anhydrous K₂CO₃) gave δ 2.12 (s, Ac), 2.88 (s, 2-Me), 6.24 (s, OCH₂O), 7.10 (s) and 7.23 (s) (ArH's and CHCl₃), and 9.12 (s, 4-H), *i.e.* only the monomer is present. Because of the very low solubility of compound (1d) no spectrum could be obtained in [²H₆]acetone.

(c) The spectrum of the benzamide (1e) in CDCl₃ is given in ref. 4 and is consistent with the exclusive presence of the monomeric form in this solvent. The intensities of the signals in the [²H₆]acetone spectrum of compound (1e) are, because of its slight solubility, very low, but all the signals of the CDCl₃ spectrum (and, except for the very intense signals of the solvent, only these) could be recognized in the [²H₆]acetone spectrum as well.

Irradiation of Compounds (1a–h) in Acetone.—The title compounds (1a–h) (1.7–11 mmol) were irradiated in dry acetone (27–87 cm³ for 1 mmol of the starting compound) with a high-pressure mercury immersion lamp (HPK-125) through Pyrex under argon at room temperature until, by t.l.c. (Kieselgel G; benzene–methanol 10 : 1 or benzene–acetone 1 : 1; detection: iodine vapour or 254 + 366 nm u.v. light) the starting compounds had been completely used up (2–50 h; see Table 1). In those cases where the starting compound did not dissolve completely, the suspensions were agitated with a magnetic stirrer until, after irradiation periods of 2–3 h, clear solutions were obtained. The initially light yellow solutions gradually turned dark.

If the resulting mixtures contained appreciable amounts of a precipitate this was filtered off and washed with small amounts of acetone and diethyl ether (fraction A). The filtrates were concentrated under reduced pressure to *ca.* 50 cm³, Kieselgel 60 (0.063–0.2 mm; Merck; 3 g) was added, and the solvent was evaporated. The residues were placed on Kieselgel columns (60–120 g, depending on the size of the

* Multiplicities from the off-resonance spectrum.

Table 4. Atomic co-ordinates of compound (11b) with e.s.d.s in parentheses

| | <i>x/a</i> | <i>y/b</i> | <i>z/c</i> | <i>B</i> _{EQ} | | <i>x/a</i> | <i>y/b</i> | <i>z/c</i> | <i>B</i> _{EQ} |
|-------|-------------|------------|------------|------------------------|--------|-------------|------------|------------|------------------------|
| C(1) | 0.260 5(5) | 1.937 8(4) | 0.178 4(2) | 3.87(31) | C(40) | -0.252 9(6) | 1.878 2(5) | 0.439 9(4) | 7.39(49) |
| O(2) | 0.299 1(3) | 1.872 1(2) | 0.109 4(2) | 4.05(21) | C(1B) | 0.653 1(8) | 0.660 5(7) | 0.198 8(4) | 11.88(76) |
| C(3) | 0.299 5(4) | 1.737 7(3) | 0.125 9(2) | 3.00(26) | C(2B) | 0.697 0(7) | 0.751 3(8) | 0.233 9(4) | 11.11(74) |
| C(4) | 0.228 4(4) | 1.731 9(3) | 0.196 6(2) | 3.03(27) | C(3B) | 0.795 3(8) | 0.804 8(7) | 0.200 4(4) | 8.96(67) |
| O(5) | 0.184 6(3) | 1.862 8(2) | 0.225 9(1) | 4.01(20) | C(4B) | 0.857 4(7) | 0.771 5(8) | 0.133 5(4) | 9.77(71) |
| C(6) | 0.353 9(4) | 1.625 8(4) | 0.083 1(2) | 3.19(28) | C(5B) | 0.824 3(8) | 0.679 9(8) | 0.097 5(4) | 10.65(78) |
| C(7) | 0.336 0(4) | 1.498 5(3) | 0.113 6(2) | 2.59(25) | C(6B) | 0.723 9(9) | 0.624 1(7) | 0.130 9(5) | 11.58(76) |
| C(8) | 0.261 9(4) | 1.494 6(3) | 0.183 9(2) | 2.35(23) | H(1A) | 0.1934 | 2.0547 | 0.1660 | 4.00 |
| C(9) | 0.207 8(4) | 1.615 3(3) | 0.227 3(2) | 2.69(25) | H(1B) | 0.3599 | 1.9347 | 0.2089 | 4.00 |
| N(10) | 0.397 7(3) | 1.381 6(3) | 0.072 3(2) | 3.11(22) | H(6) | 0.4160 | 1.6094 | 0.0332 | 4.00 |
| C(11) | 0.379 7(4) | 1.265 3(3) | 0.101 6(2) | 2.93(26) | H(9) | 0.1660 | 1.6094 | 0.2832 | 4.00 |
| N(12) | 0.301 6(3) | 1.251 1(3) | 0.166 7(2) | 2.56(20) | H(N) | 0.1094 | 1.4434 | 0.2988 | 4.00 |
| C(13) | 0.245 4(4) | 1.363 0(3) | 0.206 8(2) | 2.26(23) | H(14A) | 0.5000 | 1.1387 | 0.0156 | 4.00 |
| C(14) | 0.449 2(5) | 1.135 2(4) | 0.059 1(2) | 4.41(32) | H(14B) | 0.3594 | 1.1094 | 0.0488 | 4.00 |
| N(15) | 0.167 1(3) | 1.353 6(3) | 0.273 1(2) | 2.60(20) | H(14C) | 0.5000 | 1.0547 | 0.0820 | 4.00 |
| C(16) | 0.161 3(4) | 1.226 5(3) | 0.310 4(2) | 2.10(22) | H(18) | -0.0840 | 1.1094 | 0.4160 | 4.00 |
| C(17) | 0.016 9(3) | 1.232 1(3) | 0.340 4(2) | 2.21(22) | H(21) | 0.4160 | 1.0820 | 0.2988 | 4.00 |
| C(18) | 0.008 7(4) | 1.112 5(3) | 0.383 9(2) | 2.79(25) | H(23A) | 0.3594 | 0.6934 | 0.4492 | 4.00 |
| C(19) | 0.144 8(4) | 0.992 4(3) | 0.392 4(2) | 2.95(26) | H(23B) | 0.3320 | 0.6934 | 0.3652 | 4.00 |
| C(20) | 0.285 2(4) | 0.987 5(3) | 0.360 9(2) | 2.98(26) | H(25) | -0.1113 | 1.4160 | 0.2832 | 4.00 |
| C(21) | 0.300 1(4) | 1.102 9(3) | 0.320 2(2) | 2.71(25) | H(33A) | -0.2793 | 1.1387 | 0.1484 | 4.00 |
| O(22) | 0.166 6(3) | 0.863 6(3) | 0.432 4(2) | 4.94(22) | H(33B) | -0.1860 | 1.1070 | 0.1042 | 4.00 |
| C(23) | 0.321 8(5) | 0.767 6(4) | 0.410 7(3) | 5.47(37) | H(34A) | -0.1406 | 1.3320 | 0.0664 | 4.00 |
| O(24) | 0.402 9(3) | 0.854 0(3) | 0.378 2(2) | 4.67(22) | H(34B) | 0.0260 | 1.1939 | 0.1013 | 4.00 |
| C(25) | -0.134 0(4) | 1.364 6(3) | 0.325 6(2) | 2.55(24) | H(34C) | -0.0821 | 1.1665 | 0.0320 | 4.00 |
| N(26) | -0.194 8(3) | 1.454 3(3) | 0.389 6(2) | 2.63(20) | H(35A) | -0.5000 | 1.2500 | 0.3984 | 4.00 |
| N(27) | -0.336 7(3) | 1.453 4(3) | 0.420 9(2) | 3.06(22) | H(35B) | -0.6113 | 1.3887 | 0.3496 | 4.00 |
| C(28) | -0.377 7(4) | 1.381 4(3) | 0.379 3(2) | 3.08(26) | H(35C) | -0.5840 | 1.3887 | 0.4492 | 4.00 |
| N(29) | -0.270 5(3) | 1.330 0(3) | 0.319 4(2) | 2.95(21) | H(39A) | -0.0293 | 1.6934 | 0.4648 | 4.00 |
| C(30) | -0.289 3(4) | 1.263 1(4) | 0.261 8(2) | 3.99(32) | H(39B) | -0.1823 | 1.7409 | 0.5350 | 4.00 |
| O(31) | -0.388 0(3) | 1.217 5(3) | 0.260 8(2) | 6.13(27) | H(40A) | -0.1953 | 1.9434 | 0.4492 | 4.00 |
| O(32) | -0.180 8(3) | 1.255 6(3) | 0.207 9(2) | 4.73(24) | H(40B) | -0.2227 | 1.8594 | 0.3828 | 4.00 |
| C(33) | -0.187 6(6) | 1.187 4(6) | 0.143 2(3) | 7.76(47) | H(40C) | -0.3744 | 1.9197 | 0.4519 | 4.00 |
| C(34) | -0.093 4(8) | 1.221 1(8) | 0.083 0(3) | 10.46(75) | H(1B) | 0.5742 | 0.6239 | 0.2220 | 4.00 |
| C(35) | -0.523 0(4) | 1.356 4(4) | 0.394 4(3) | 4.20(32) | H(2B) | 0.6483 | 0.7763 | 0.2880 | 4.00 |
| C(36) | -0.136 7(4) | 1.548 7(3) | 0.405 6(2) | 2.86(26) | H(3B) | 0.8207 | 0.8800 | 0.2291 | 4.00 |
| O(37) | -0.019 0(3) | 1.555 3(2) | 0.373 6(1) | 3.22(18) | H(4B) | 0.9402 | 0.8146 | 0.1064 | 4.00 |
| O(38) | -0.219 6(3) | 1.629 1(2) | 0.459 7(1) | 3.39(19) | H(5B) | 0.8795 | 0.6537 | 0.0407 | 4.00 |
| C(39) | -0.165 1(4) | 1.732 8(4) | 0.482 8(2) | 3.77(30) | H(6B) | 0.7040 | 0.5514 | 0.0963 | 4.00 |

Table 5. Bond lengths (Å) between the non-hydrogen atoms of compound (11b) with e.s.d.s in parentheses

| | | | | | | | |
|-------------|----------|-------------|----------|-------------|----------|-------------|-----------|
| C(1)-O(2) | 1.431(6) | C(11)-N(12) | 1.358(5) | C(20)-O(24) | 1.378(5) | O(32)-C(33) | 1.442(8) |
| C(1)-O(5) | 1.425(6) | C(11)-C(14) | 1.489(7) | O(22)-C(23) | 1.429(5) | C(33)-C(34) | 1.444(11) |
| O(2)-C(3) | 1.374(6) | N(12)-C(13) | 1.318(5) | C(23)-O(24) | 1.436(7) | C(36)-O(37) | 1.222(5) |
| C(3)-C(4) | 1.398(6) | C(13)-N(15) | 1.368(5) | C(25)-N(26) | 1.483(5) | C(36)-O(38) | 1.328(5) |
| C(3)-C(6) | 1.349(7) | N(15)-C(16) | 1.418(6) | C(25)-N(29) | 1.469(5) | O(38)-C(39) | 1.459(6) |
| C(4)-O(5) | 1.383(6) | C(16)-C(17) | 1.392(5) | N(26)-N(27) | 1.400(4) | C(39)-C(40) | 1.528(9) |
| C(4)-C(9) | 1.341(7) | C(16)-C(21) | 1.396(5) | N(26)-C(36) | 1.351(6) | C(1B)-C(2B) | 1.383(14) |
| C(6)-C(7) | 1.427(7) | C(17)-C(18) | 1.408(6) | N(27)-C(28) | 1.284(6) | C(1B)-C(6B) | 1.359(13) |
| C(7)-C(8) | 1.402(6) | C(17)-C(25) | 1.518(5) | C(28)-N(29) | 1.402(5) | C(2B)-C(3B) | 1.326(12) |
| C(7)-N(10) | 1.370(6) | C(18)-C(19) | 1.361(5) | C(28)-C(35) | 1.479(6) | C(3B)-C(4B) | 1.315(12) |
| C(8)-C(9) | 1.425(6) | C(19)-C(20) | 1.370(5) | N(29)-C(30) | 1.363(7) | C(4B)-C(5B) | 1.340(14) |
| C(8)-C(13) | 1.431(6) | C(19)-O(22) | 1.378(6) | C(30)-O(31) | 1.192(6) | C(5B)-C(6B) | 1.358(13) |
| N(10)-C(11) | 1.319(6) | C(20)-C(21) | 1.372(7) | C(30)-O(32) | 1.331(6) | | |

run) and chromatographed initially with benzene and subsequently with benzene-acetone mixtures in which the proportion of the more polar component was gradually increased until a 1:1 ratio was reached. 10-cm³ Fractions were collected and those containing identical components were combined and evaporated to dryness under reduced pressure; the residues (fractions B) were crystallized from the appropriate solvents.

Those fractions B which failed to crystallize were further purified by t.l.c. (Kieselgel PF₂₅₄₊₃₆₆, Merck; benzene-acetone 10:1-1:1, acetone, benzene-methanol, 10:1-

5:1, dichloromethane-acetone 7:3-7:1, respectively; detection: 254 + 366 u.v. light) to obtain the fractions C. The filtrates of the fractions A of the smaller runs were similarly worked up. Products and yields are listed in Table 1.

Known compounds [(4a),⁶ (6b),⁶ (7f)≡(7a),¹⁰ (13a-c),⁹ and those listed in Table 2] were identified by comparison with authentic samples. The following new compounds were obtained: 3-acetylamino-2-methyl-6,7-methylenedioxyquinazolin-4(3H)-one (7d), m.p. 246-247 °C (from EtOH) [Found: C, 54.95; H, 4.35; N, 15.9. C₁₂H₁₁N₃O₄ (M, 261.23) requires C, 55.17; H, 4.24; N, 16.09%], *v*_{max}. (KBr) 3 200,

Table 6. Bond angles (°) from the non-hydrogen atoms of compound (11b) with e.s.d.s in parentheses

| | | | | | |
|-------------------|----------|-------------------|----------|-------------------|-----------|
| O(2)-C(1)-O(5) | 106.8(6) | N(12)-C(13)-N(15) | 118.8(6) | N(27)-N(26)-C(36) | 123.2(6) |
| C(1)-O(2)-C(3) | 105.4(6) | C(13)-N(15)-C(16) | 126.1(6) | N(26)-N(27)-C(28) | 106.4(6) |
| O(2)-C(3)-C(4) | 109.0(6) | N(15)-C(16)-C(17) | 118.3(6) | N(27)-C(28)-N(29) | 112.9(6) |
| O(2)-C(3)-C(6) | 128.3(7) | N(15)-C(16)-C(21) | 119.8(6) | N(27)-C(28)-C(35) | 122.6(7) |
| C(4)-C(3)-C(6) | 122.7(7) | C(17)-C(16)-C(21) | 121.9(6) | N(29)-C(28)-C(35) | 124.5(7) |
| C(3)-C(4)-O(5) | 108.8(6) | C(16)-C(17)-C(18) | 120.0(6) | C(25)-N(29)-C(28) | 109.5(6) |
| C(3)-C(4)-C(9) | 122.9(7) | C(16)-C(17)-C(25) | 121.1(6) | C(25)-N(29)-C(30) | 123.9(6) |
| O(5)-C(4)-C(9) | 128.2(7) | C(18)-C(17)-C(25) | 118.9(6) | C(28)-N(29)-C(30) | 126.6(6) |
| C(1)-O(5)-C(4) | 105.4(6) | C(17)-C(18)-C(19) | 117.3(7) | N(29)-C(30)-O(31) | 124.4(8) |
| C(3)-C(6)-C(7) | 116.6(7) | C(18)-C(19)-C(20) | 121.9(7) | N(29)-C(30)-O(32) | 109.9(7) |
| C(6)-C(7)-C(8) | 120.3(7) | C(18)-C(19)-O(22) | 128.0(7) | O(31)-C(30)-O(32) | 125.7(8) |
| C(6)-C(7)-N(10) | 117.2(6) | C(20)-C(19)-O(22) | 110.1(6) | C(30)-O(32)-C(33) | 115.7(7) |
| C(8)-C(7)-N(10) | 122.4(6) | C(19)-C(20)-C(21) | 122.9(7) | O(32)-C(33)-C(34) | 108.9(10) |
| C(7)-C(8)-C(9) | 120.9(6) | C(19)-C(20)-O(24) | 109.7(6) | N(26)-C(36)-O(37) | 122.5(7) |
| C(7)-C(8)-C(13) | 114.9(6) | C(21)-C(20)-O(24) | 127.4(7) | N(26)-C(36)-O(38) | 112.5(6) |
| C(9)-C(8)-C(13) | 124.2(6) | C(16)-C(21)-C(20) | 116.0(6) | O(37)-C(36)-O(38) | 125.0(7) |
| C(4)-C(9)-C(8) | 116.6(7) | C(19)-O(22)-C(23) | 104.6(6) | C(36)-O(38)-C(39) | 117.2(6) |
| C(7)-N(10)-C(11) | 116.1(6) | O(22)-C(23)-O(24) | 107.3(7) | O(38)-C(39)-C(40) | 108.8(7) |
| N(10)-C(11)-N(12) | 127.0(7) | C(20)-O(24)-C(23) | 104.7(6) | C(2B)-C(1B)-C(6B) | 115.8(14) |
| N(10)-C(11)-C(14) | 117.4(7) | C(17)-C(25)-N(26) | 114.0(6) | C(1B)-C(2B)-C(3B) | 121.0(14) |
| N(12)-C(11)-C(14) | 115.6(7) | C(17)-C(25)-N(29) | 113.0(6) | C(2B)-C(3B)-C(4B) | 121.6(14) |
| C(11)-N(12)-C(13) | 116.5(6) | N(26)-C(25)-N(29) | 97.8(5) | C(3B)-C(4B)-C(5B) | 120.4(15) |
| C(8)-C(13)-N(12) | 122.9(6) | C(25)-N(26)-N(27) | 112.9(5) | C(4B)-C(5B)-C(6B) | 118.9(15) |
| C(8)-C(13)-N(15) | 118.3(6) | C(25)-N(26)-C(36) | 122.6(6) | C(1B)-C(6B)-C(5B) | 122.2(15) |

1 705, and 1 670 cm^{-1} ; δ 2.22 (s, Ac), 2.48 (s, 2-Me), 6.03 (s, OCH_2O), 6.95 (s, 8-H), 7.42 (s, 5-H), and 8.1 (bs, NH); m/z (150 °C) (% rel. int.) 261 (80, M^+), 219 (100), 204 (3.1), 203 (5.2), 202 (3.7), 190 (75), 161 (6.6), 120 (25), and 119 (8.9).

3-Benzoylamino-2-methyl-6,7-methylenedioxyquinazolin-4(3H)-one (7h)≡(7e) was identified by comparison with an authentic sample, see below.

4-(2-Methyl-6,7-methylenedioxyquinazolin-4-yl)butan-2-one (9h), m.p. 149–151 °C (from MeOH) [Found: C, 65.1; H, 5.4; N, 10.7. $\text{C}_{14}\text{H}_{14}\text{N}_2\text{O}_3$ (M , 258.27) requires C, 65.10; H, 5.46; N, 10.85%]; ν_{max} (KBr) 1 730 cm^{-1} ; δ 2.28 (s, Ac), 2.70 (s, 2-Me), 3.01 (t) and 3.36 (t) (CH_2CH_2), 6.06 (s, OCH_2O), and 7.12 (s) and 7.23 (s) (ArH); m/z (80 °C) (% rel. int.) 257 (8.5, M^+), 243 (13), 215 (100), 202 (4.5), 188 (3.5), 187 (7.0), 185 (6.7), and 157 (12). Compound (9h) was not detected (t.l.c.) in a mixture resulting from irradiation for 80 h of compound (4h) (2.45 mmol) in dry acetone (150 cm^3); only tarry products were formed under these conditions.

Diethyl 5-[4,5-methylenedioxy-2-(6,7-methylenedioxyquinazolin-4-ylamino)phenyl]- Δ^2 -1,2,4-triazoline-1,4-dicarboxylate (11a), m.p. 167–169 °C (from benzene–light petroleum) [Found: C, 55.35; H, 4.35; N, 15.95. $\text{C}_{24}\text{H}_{22}\text{N}_6\text{O}_8$ (M , 522.47) requires C, 55.17; H, 4.25; N, 16.09%]; ν_{max} (KBr) 3 400 and 1 730 cm^{-1} ; δ (CDBr₃; 90 °C) * 1.18 (t), 1.37 (t), 4.18 (q), and 4.33 (q) (2 CO_2Et groups), 6.02 (s) and 6.13 (s) (2 OCH_2O groups), 6.68 (s), 6.82 (s), 7.20 (s), 7.32 (s), 7.33 (s), and 7.42 (s) (4 ArH, 3- and 5-H), 8.49 (s, 2-H, quinazoline ring), and 8.87 (bs, NH); m/z (200 °C) (% rel. int.) 522 (5.0, M^+), 449 (36), 434 (12), 408 (31), 407 (46), 406 (36), 377 (21), 361 (21), 335 (14), 334 (20), 320 (100), 308 (22), 262 (8.7), 189 (11), 175 (6.2), 174 (10), and 173 (11).

Diethyl 3-methyl-5-[4,5-methylenedioxy-2-(2-methyl-6,7-methylenedioxyquinazolin-4-ylamino)phenyl]- Δ^2 -1,2,4-triazoline-1,4-dicarboxylate (11b), m.p. 218 °C (from benzene–light petroleum) [Found: C, 56.85; H, 4.75; N, 15.15. $\text{C}_{26}\text{H}_{26}\text{N}_6\text{O}_8$ (M , 550.49) requires C, 56.72; H, 4.76; N, 15.27%]; ν_{max} (KBr) 3 300 and 1 750 cm^{-1} ; δ (CDBr₃; 90 °C †) 0.97 (t), 1.35 (t), 3.95 (m), and 4.30 (q) (2 CO_2Et groups), 2.42 (s) and

2.50 (s) (2 Me), 5.96 and 6.01 (two inner peaks of an AB spectrum, OCH_2O), 6.07 (s, OCH_2O), 6.58 (s), 6.78 (merged with CHBr_3 signal), 7.11 (s), 7.21 (s), and 7.29 (s) (4 ArH and 5-H), and 8.90 (bs, NH); m/z (200 °C) (% rel. int.) 550 (12, M^+), 477 (100), 462 (4.2), 433 (8.7), 421 (8.0), 419 (6.9), 405 (16), 375 (7.0), 334 (13), 322 (16.2), 203 (6.2), and 187 (15).

Dibenzyl 3-methyl-5-[4,5-methylenedioxy-2-(2-methyl-6,7-methylenedioxyquinazolin-4-ylamino)phenyl]- Δ^2 -1,2,4-triazoline-1,4-dicarboxylate (11c), m.p. 135–136 °C (from MeOH) [Found: C, 63.9; H, 4.5; N, 12.6. $\text{C}_{36}\text{H}_{30}\text{N}_6\text{O}_8$ (M , 674.65) requires C, 64.08; H, 4.48; N, 12.46%]; ν_{max} (KBr) 3 350 and 1 745 cm^{-1} ; δ (CDBr₃; 80 °C) 2.43 (s) and 2.52 (s) (2 Me), 4.87, 5.04 and 5.18, 5.33 (two AB spectra, J 12.5 and 12.6 Hz, respectively, 2 PhCH_2O), 6.0 (s) and 6.15 (s) (2 OCH_2O groups), 6.50 (s, 1 H) and 6.8–7.45 (m, 14 H) (2 Ph, 4 ArH, and 5-H), and 9.0 (bs, NH); m/z (250 °C) (% rel. int.) 674 (0.8, M^+), 539 (11), 495 (9.4), 430 (23), 429 (15), 404 (7.2), 334 (28), 322 (4.3), 187 (24), 108 (18), 107 (14), and 91 (100).

1-Benzoylimino-1-(2-methyl-6,7-methylenedioxyquinazolin-4-yl)acetone (12e), identical (m.p., mixed m.p., i.r.) with an authentic sample, see below.

Irradiation of Compounds (1) in Various Solvents.—Irradiation and work-up of the resulting mixtures were carried out exactly as described above for irradiation in acetone. For details, see Table 3. The only new compound obtained in these experiments was ethyl 2-methyl-7,8-methylenedioxy-5-*t*-butoxy-4,5-dihydro-3H-1,3,4-benzotriazepine-4-carboxylate (15), m.p. 145–146 °C (from benzene–light petroleum) [Found: C, 58.2; H, 6.7; N, 12.3. $\text{C}_{17}\text{H}_{23}\text{N}_3\text{O}_5$ (M , 349.37) requires C, 58.44; H, 6.63; N, 12.03%]; δ 1.25 (s, Bu^t), 1.28 (t) and 4.18 (q) (CO_2Et), 2.12 (s, 2-Me), 5.83 (s, OCH_2O), 6.24 (s), 6.37 (s), and 6.56 (s) (5-, 6-, and 9-H), and 6.7 (bs, NH); m/z (140 °C) (% rel. int.) 349 (22, M^+), 292 (4.2), 276 (36), 264 (8.8), 205 (8.2), 190 (100), 162 (12), 120 (2.8), and 104 (30).

* At room temperature in CDCl_3 the two CO_2Et groups give two unresolved broad singlets at δ 1.28 and 4.25, respectively.

† At room temperature in CDCl_3 the two CO_2Et groups give poorly resolved triplets (δ 0.95 and 1.35) and quadruplets (3.97 and 4.32), respectively.

N-[2-Methyl-6,7-methylenedioxy-4-(2-oxopropylidene)-3,4-dihydroquinazolin-3-yl]benzamide (17a).—A solution of the benzamide (13e)⁹ (0.5 g, 1.36 mmol) in dichloromethane-dioxane-water (4 cm³ each) was cooled to -5 °C. An aqueous solution (8 cm³) of K₃[Fe(CN)₆] (0.95 g, 2.9 mmol) and, subsequently, an aqueous (2.5 cm³) solution of KOH (0.42 g, 7.5 mmol) were added dropwise with continuous stirring at such a rate that the temperature did not exceed -5 °C. The solution was allowed to warm to 0 °C whereupon it turned dark brown. The mixture was extracted with dichloromethane (3 × 20 cm³), dried (MgSO₄) and evaporated to dryness. The residue was worked up by preparative t.l.c. (Kieselgel PF₂₅₄₊₃₆₆, benzene-acetone 1 : 1) to give yellow crystals (0.25 g, 51%) of the title compound, m.p. 196–197 °C (from benzene) [Found: C, 66.25; H, 4.65; N, 11.65. Calc. for C₂₀H₁₇N₃O₄ (*M*, 363.36): C, 66.10; H, 4.74; N, 11.56%; *v*_{max} (KBr) 3 350, 1 690, and 1 640 cm⁻¹; *δ* 2.20 (s, Ac), 2.59 (s, 2-Me), 5.90 (s, CH=), 6.09 (s, OCH₂O), 7.02 (s), and 7.27 (s) (ArH, quinazoline ring), 7.40–7.58 (m, *m*- and *p*-H, benzoyl), 7.90–8.05 (m, *o*-H, benzoyl), and 13.72 (bs, NH); *m/z* (150 °C) (% rel. int.) 363 (22, *M*⁺), 348 (4.3), 346 (4.0), 306 (19), 304 (6.6), 258 (4.8), 241 (13), 218 (18), 217 (24), 188 (7.9), 187 (7.2), 161 (5.3), 105 (100), and 77 (24).

1-Benzoylimino-1-(2-methyl-6,7-methylenedioxyquinazolin-4-yl)acetone (12e).—Compound (17a) (0.48 g, 1.32 mmol) in dry acetone (150 cm³) was irradiated with a high-pressure mercury immersion lamp (HPK-125) through Pyrex at room temperature under argon for 35 h. The bright yellow solution was evaporated to dryness and the residue was worked up by preparative t.l.c. (Kieselgel PF₂₅₄₊₃₆₆, benzene-acetone, 10 : 1) to give the title compound (12.2 mg, 2.6%), m.p. 218–219 °C (from MeOH) as colourless crystals [Found: C, 66.6; H, 4.25; N, 11.6. Calc. for C₂₀H₁₅N₃O₄ (*M*, 361.34): C, 66.47; H, 4.18; N, 11.63%; *v*_{max} (KBr) 1 760 and 1 615 cm⁻¹; *δ* 2.44 (s, Ac), 2.94 (s, 2-Me, quinazoline ring), 6.04 (s, OCH₂O), 7.19 (s) and 7.21 (s) (ArH, quinazoline ring), 7.35–7.45 (m, *m*- and *p*-H, benzoyl), and 7.75–7.90 (m, *o*-H, benzoyl); *m/z* (70 °C) (% rel. int.) 361 (16, *M*⁺), 319 (100), 188 (11), 187 (34), 147 (7.0), 146 (4.3), 90 (11), 88 (8.6), and 43 (6.9).

3-Benzoylamino-2-methyl-6,7-methylenedioxyquinazolin-4(3H)-one (7e).—The solution of compound (13e)⁹ (1.0 g, 2.74 mmol) in dry acetone (150 cm³) was irradiated with a high-pressure mercury immersion lamp (HPH-125) through Pyrex for 50 h under oxygen at room temperature. The mixture was evaporated to dryness and the residue which contained substantial amounts of tarry products was worked up by t.l.c. (Kieselgel PF₂₅₄₊₃₆₆, benzene-acetone, 1 : 1) to obtain traces of compound (17a) and compound (7e)·0.5MeOH (0.37 g, 42%), m.p. 210–211 °C (from MeOH) [Found: C, 61.8; H, 4.25; N, 12.45. Calc. for 2 C₁₇H₁₃N₃O₄·MeOH (*M*, 678.64): C, 61.94; H, 4.46; N, 12.38%; *v*_{max} (KBr) 3 600–2 800, and 1 690b cm⁻¹; *δ* 2.48 (2-Me), 3.42 (0.5 MeOH), 6.03 (s, OCH₂-

O), 6.91 (s, 8 H), 7.34 (s, 5-H), 7.20–7.50 (m, *m*- and *p*-H, benzoyl), 7.80–7.92m (*o*-H, benzoyl), and 9.7 (s, NH); *m/z* (160 °C) (% rel. int.) 323 (24), 188 (1.4), 161 (1.5), 120 (7.3), 119 (3.4), 105 (100), 77 (23), 62 (2.3), and 51 (3.2).

X-Ray Structure Determination of Compound (11b).—*Crystal data*. C₂₆H₂₆N₆O₈·C₆H₆, *M* = 628.7, *a* = 9.324(1), *b* = 10.235(1), *c* = 18.041(3) Å, *α* = 83.80(1)° *β* = 85.52(1)°, *γ* = 65.77(1)°, *V* = 1559.7(4) Å³ (from single crystal diffractometry), space group P1̄ (from structure refinement), *Z* = 2, *d*_x = 1.338 g cm⁻³, *μ*(Mo-K_α; λ 0.710 73 Å) = 0.92 cm⁻¹. 5 482 Reflections were collected on an Enraf-Nonius CAD-4 computer-controlled automatic four-circle diffractometer with monochromated Mo-K_α radiation. The structure was solved by direct methods¹² and Fourier techniques. The solvent molecule and the hydrogen atoms were located in difference maps. The structure was refined by anisotropic least-squares to a final *R* = 0.057 for 3 280 reflections [*F*_o ≥ 5.0σ(*F*_o)]. No absorption correction was applied. All calculations were performed on a PDP-11/34 64k minicomputer using the E.N. SDP program package and local programs.

Acknowledgements

We thank Dr. Á. Szöllősy and staff for the n.m.r. spectra, Dr. K. Kiss-Erős and staff for the i.r. spectra, and Dr. I. Balogh-Batta for the microanalyses.

References

- 1 Part 23, J. Fetter, K. Lempert, J. Møller, J. Nyitrai, and K. Zauer, *Acta Chim. Hung.*, 1983, **112**, 43.
- 2 Part 10: F. Bertha, J. Fetter, K. Lempert, J. Møller, and L. Radics, *J. Chem. Res.*, 1980, (S), 402; (M) 4737.
- 3 K. Lempert, *Lect. Heterocycl. Chem.*, 1982, **6**, S-25.
- 4 J. Fetter, K. Lempert, and J. Møller, *Tetrahedron*, 1975, **31**, 2559.
- 5 L. Párkányi, Gy. Argay, J. Fetter, and K. Lempert, *Cryst. Struct. Commun.*, 1977, **6**, 137.
- 6 J. Fetter, K. Lempert, G. Barta-Szalai, J. Møller, and L. Párkányi, *Acta Chim. Acad. Sci. Hung.*, 1977, **94**, 233.
- 7 G. Barta-Szalai, J. Fekete, J. Fetter, K. Lempert, and J. Møller, *Acta Chem. Scand., Ser. B*, 1979, **33**, 79.
- 8 G. Barta-Szalai, J. Fetter, K. Lempert, and J. Møller, *Acta Chim. Acad. Sci. Hung.*, 1980, **104**, 253.
- 9 G. Barta-Szalai, I. Csöregy, M. Czugler, J. Fetter, K. Lempert, and J. Møller, *J. Chem. Res.*, 1981, (S), 269; (M), 3301.
- 10 J. Fetter, G. Barta-Szalai, A. Jaber, and F. Bertha, *Periodica Polytech., Chem. Eng.*, 1978, **22**, 143.
- 11 J. Fetter, K. Lempert, and J. Møller, *Acta Chim. Acad. Sci. Hung.*, 1976, **88**, 435.
- 12 G. Germain, P. Main, and M. M. Woolfson, *Acta Crystallogr., Sect. A*, 1971, **27**, 368.

Received 10th December 1982; Paper 2/2069