Synthesis of 5*H*,12*H*-Quinazolino[3,2-*a*][3,1]benzoxazine-5,12-diones

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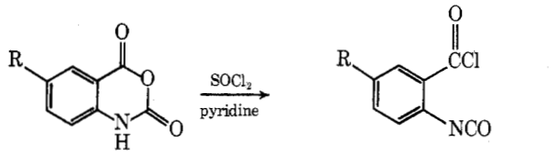
A coproduct of 5-chloro-2-isocyanatobenzoyl chloride (**2b**) produced in the reaction of 5-chloroisatoic anhydride (**1b**) with thionyl chloride and a catalytic amount of pyridine has been identified as 3,10-dichloro-5*H*,12*H*-quinazolino[3,2-*a*][3,1]benzoxazine-5,12-dione (**3**). As a result, a new synthesis of the tetracyclic ring system represented by **3**, from isatoic anhydrides and *o*-cyanatobenzoyl chlorides, was developed. The mechanism for the formation of quinazolinobenzoxazinediones by this novel method is discussed.

We recently reported[1] the use of 2-isocyanatobenzoyl chloride (**2a**) and its-5-chloro analogue (**2b**) in the preparation of 3,4-dihydro-4-methyl-1*H*-1,3,4-benzotriazepine-2,5-diones. The reaction of isatoic anhydride (**1a**) with thionyl chloride in the presence of a catalytic amount of pyridine[2] yielded a solution which, after 24 h, was concentrated and distilled to afford 82% of **2a**. Treatment of 5-chloroisatoic anhydride (**1b**) with thionyl chloride in the presence of a catalytic amount of pyridine yielded a solution only after additional thionyl chloride and dioxane had been added, and reflux had been maintained for 3 weeks.[1] Concentration of this solution yielded a yellow liquid, which was distilled to afford 5-chloro-2-isocyanatobenzoyl chloride (**2b**) in 31% yield, and a yellow solid (Scheme I).

Scheme I



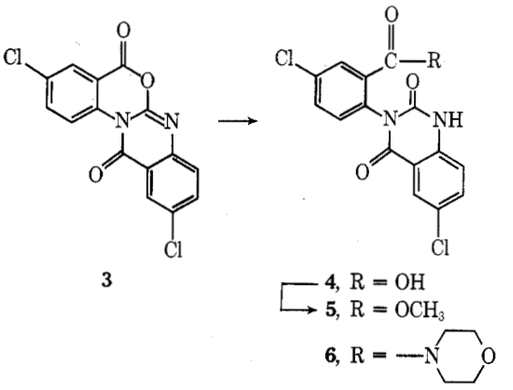
1a, R = H **2a**, R = H (82%)
b, R = Cl **b**, R = Cl (31%) + **3** (42%)

On the basis of spectral, chemical, and analytical data, we have now identified this yellow solid, which was produced in 42% yield, as 3,10-dichloro-5*H*,12*H*-quinazolino[3,2-*a*]-[3,1]benzoxazine-5,12-dione[1] (**3**). The mass spectrum (70 eV) of **3** displayed a molecular ion at *m/e* 332 and combustion analysis indicated a molecular formula of $C_{14}H_6Cl_2N_2O_3$. The NMR spectrum of **3** indicated only the presence of aromatic protons, and the infrared spectrum showed only CH stretching above 3000 cm^{-1}, and intense absorption bands at 1765, 1695, and 1620 cm^{-1}, which we assign to the benzoxazinone carbonyl, quinazolinone carbonyl, and C=N groups of **3**, respectively.

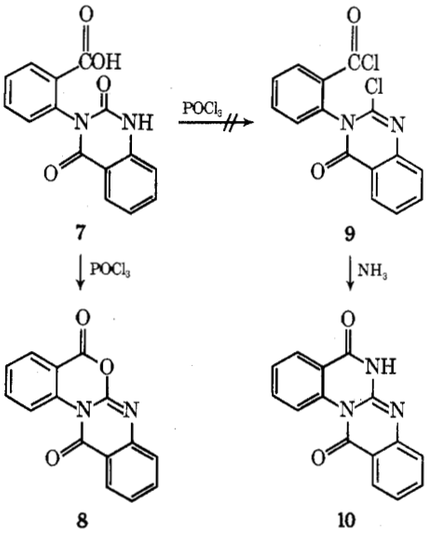
Quinazolobenzoxazinedione **3** was reactive, as would be predicted,[3] toward nucleophiles. Attempted recrystallization of **3** from dioxane produced 5-chloro-2-[6-chloro-1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl]benzoic acid (**4**), from the small amount of water present in the solvent. Treatment of **4** with 3-methyl-1-*p*-tolyltriazine (TMT) gave methyl ester **5**, which was also produced by dissolving **3** in methanol and dimethyl sulfoxide. Brief treatment of **3** with morpholine produced the morpholine amide **6**. See Scheme II.

The parent compound of the ring system represented by **3**, 5*H*,12*H*-quinazolino[3,2-*a*][3,1]benzoxazine-5,12-dione (**8**), has been reported by Doleschall and Lempert.[3] In an attempt to prepare 5*H*-quinazolino[3,2-*a*]quinazoline-5,12(6*H*)-dione (**10**), a compound isosteric with **7**, Doleschall and Lempert first treated 2-[1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl]benzoic acid (**7**) with phosphorus oxychloride.[4] These authors envisioned the production of acid chloride **9** from this reaction,

Scheme II



Scheme III

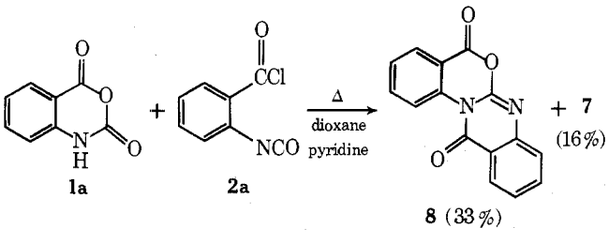


which they could then react with ammonia to give **10**,[5] and with primary amines to yield 6-substituted derivatives of **10**. Instead, cyclodehydration occurred to produce **8** rather than the expected **9** (Scheme III). The structure of **8** was confirmed and other possible isomeric products were eliminated on the basis of chemical reactivity and spectral data.

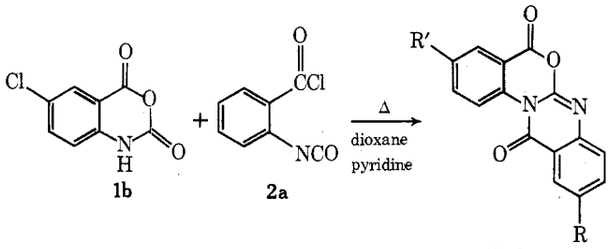
We have also prepared compound **8** by treating isatoic anhydride (**1a**) and 2-isocyanatobenzoyl chloride (**2a**) with a catalytic amount of pyridine in dioxane at reflux. Acid **7**, which was a coproduct in this reaction, was probably produced from **8** by hydrolysis during workup (Scheme IV).

Several mechanistic possibilities for the formation of **8** were considered at this point. The results shown in Scheme IV did not rule out mechanisms which could be envisioned for the

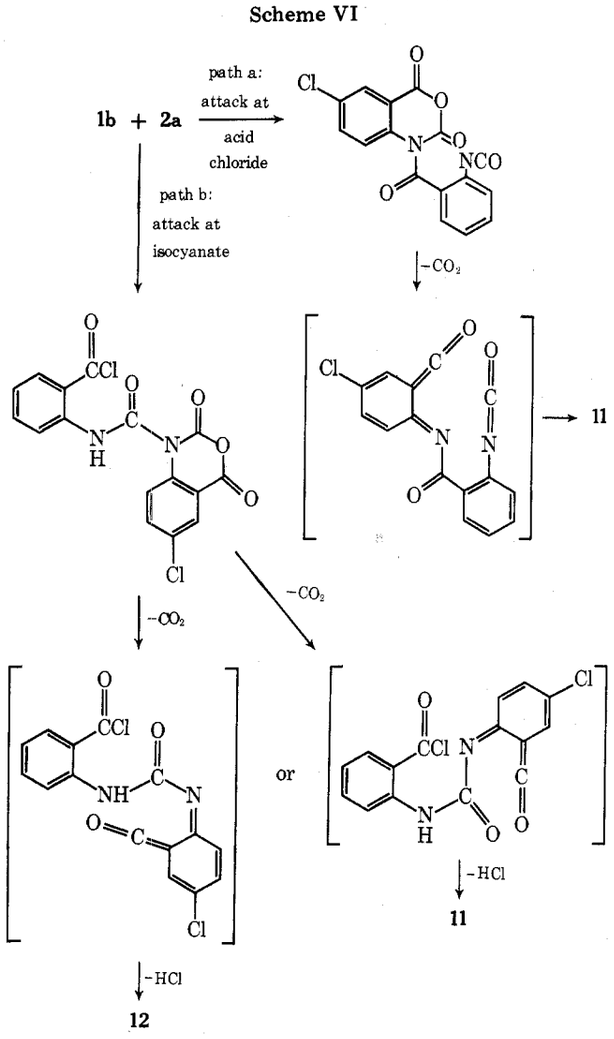
Scheme IV



Scheme V



Scheme VI

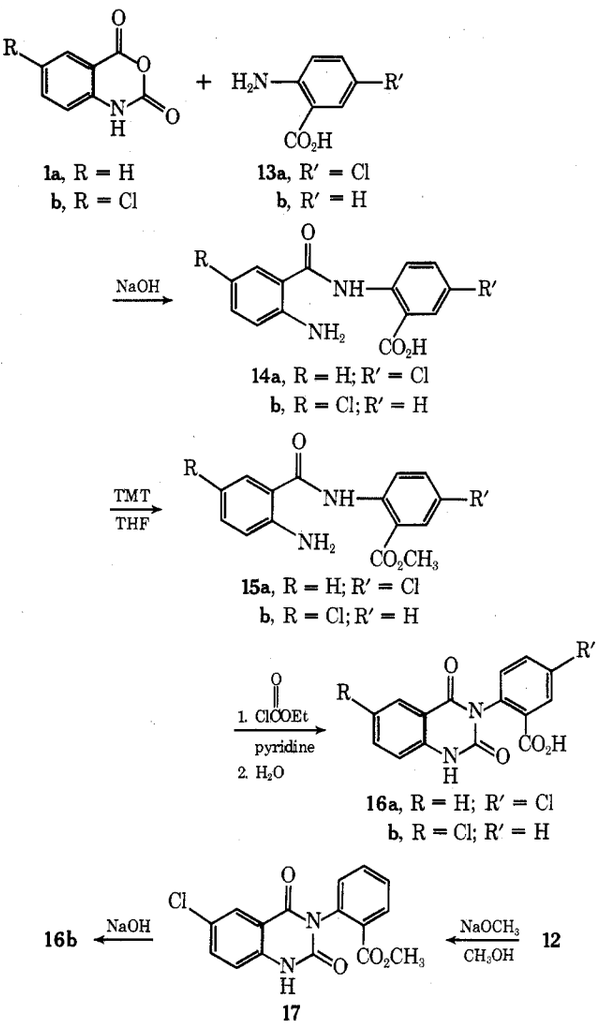


production of 8 from either **1a** alone or **2a** alone. An experiment which did rule out these possibilities, however, is shown in Scheme V. When equimolar amounts of 5-chloroisatoic anhydride (**1b**) and 2-isocyanatobenzoyl chloride (**2a**) and a catalytic amountof pyridine were heated at reflux in dioxane until **2a** was no longer present, a 60% yield of a single chloroquinazolinobenzoxazinedione, either **11** or **12**, was obtained.

Scheme VI indicates possible mechanistic pathways for the production of a chloroquinazolobenzoxazinedione by nucleophilic attack of **1b** on **2a**. Since previous work[1] indicated that the carboxylic acid chloride group in *o*-isocyanatobenzoyl chlorides was more susceptible to nucleophilic attack than the isocyanato group, we initially favored the mechanism depicted in path a.[6] Therefore, we chose to first determine, by an unequivocal synthetic route, whether compound **11** was the chloroquinazolinobenzoxazinedione produced from **1b** and **2a** (Scheme VI).

Reaction of isatoic anhydride (**1a**) with 5-chloroanthranilic acid (**13a**) produced anthraniloylanthranilic acid (**14a**). Methylation of **14a** with 3-methyl-1-*p*-tolyltriazine (TMT) produced ester **15a**. Treatment of **15a** with ethyl chloroformate in pyridine yielded, after workup, quinazolinedione **16a**.[7] Compound **16a** was compared with the product obtained when the reaction product from Scheme V was treated with sodium methoxide and then sodium hydroxide. Since these compounds were different, it was inferred that the reaction product from Scheme V was **12**, and that the ester obtained from treating **12** with sodium methoxide was **17**. See Scheme VII.

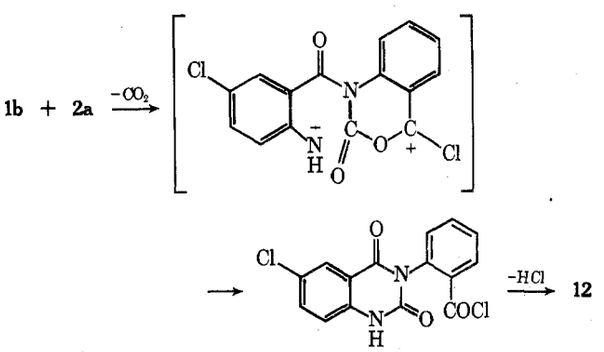
Scheme VII



To confirm these findings, an authentic sample of **16b**[7] was then prepared from 5-chloroisatoic anhydride (**1b**) and anthranilic acid (**13b**) as shown in Scheme VII. The product from these reactions was identical in all respects with the product obtained when the reaction product from Scheme V was treated with sodium methoxide and then sodium hydroxide. If the reaction of **1b** and **2a** is initiated by nucleophilic attack of the nitrogen atom of **1b**, then path b[9] (Scheme VI) must be operative.

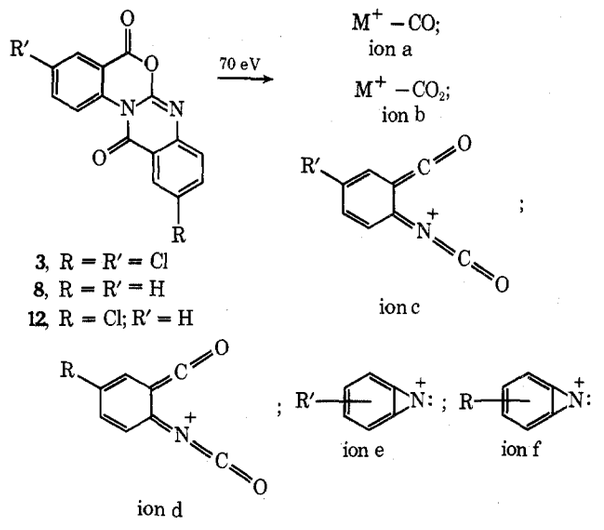
Another mechanism to be considered for the reaction of **1b** and **2a** is derived from one advanced by Staiger, Moyer, and Pitcher for the reaction of isatoic anhydride with phenyl isocyanate.[10] These authors state that "the nucleophilic nitrogen of the phenyl isocyanate attacks the number four carbon atom of isatoic anhydride, which is followed by loss of CO_2 and ring closure to the 3-phenyl-2,4-dioxo-1,2,3,4-tetrahydroquinazoline".[11] This mechanism, as applied to **1b** and **2a**, is shown in Scheme VIII. The only product predicted by this pathway is **12**, which is the observed product.

Scheme VIII



Mass spectral analysis of the product obtained from the reaction of **1b** and **2a** was not helpful in elucidating its structure. The major fragment ions observed for compounds **3**, **8**, and **12** are shown in Scheme IX. Although ions c and d are a

Scheme IX



degenerate pair for both compounds **3** and **8**, it is clear that they can arise from two different fragmentation pathways, since compound **12** produced the dissimilar ions c and d. The same is true for ions e and f. Thus, ions c, d, e, and f, all of different mass, are observed in the mass spectrum of **12**. It was, therefore, not possible to differentiate between structures **11** and **12** on the basis of mass spectral analysis.

Experimental Section[12]

3,10-Dichloro-5*H*,12*H*-quinazolino[3,2-*a*][3,1]benzoxazine-5,12-dione (3). The reaction of 5-chloroisatoic anhydride with $SOCl_2$ to produce **2b** in 31% yield and **3** in 42% yield (mp 213–218 °C) is described elsewhere.[1] Compound **3**: mp 238–242 °C (acetone); ir (Nujol) 1765 (benzoxazinone C=O), 1695 (quinazolinone C=O), 1620 cm^{-1} (C=N); NMR[13] (acetone-d_6) δ 8.44–7.17 (m, all protons); mass spectrum (70 eV) *m/e* (rel intensity) 334 (59), 332 (89), 306 (11), 304 (15), 290 (13), 288 (22), 182 (35), 180 (100), 126 (30), 124 (91).

Anal. Calcd for $C_{15}H_6Cl_2N_2O_3$: C, 54.08; H, 1.82; N, 8.41. Found: C, 54.30; H, 1.80; N, 8.66.

5-Chloro-2-[6-chloro-1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl]benzoic Acid (4). Compound **3** was quantitatively converted to **4** by crystallization from a large volume of wet dioxane. Alternatively, **3** was dissolved in wet Me_2SO to yield **4**, which was recovered by precipitation with water. Compound **4**: mp 303–305 °C: ir (Nujol) 3400–2400 (NH and OH), 1720 (acid C=O), 1670 (C=O); NMR (Me_2SO-d_6) δ 11.83 (s, 1, OH, D_2O exchangeable), 8.04–7.07 (m, 6, aromatic); mass spectrum (70 eV) *m/e* 350 (molecular ion).

Anal. Calcd for $C_{15}H_8Cl_2N_2O_4$: C, 51.31; H, 2.30; N, 7.98. Found: C, 51.63; H, 2.54; N, 7.74.

Methyl 5-Chloro-2-[6-chloro-1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl]benzoate (5). A. From 3. A 7.50-g (22.5 mmol) quantity of **3** was slurried with 75 ml of methanol and heated at reflux for 15 min. Solution had not resulted, and the ir of a concentrated aliquot of the mixture showed only starting material. A 40-ml volume of Me_2SO was added and reflux was maintained for 15 min. A small amount of insoluble material was removed by filtration. After 1 day, only a small amount of crystals had formed in the filtrate. The filtrate was warmed and the solution was diluted with water until cloudy, and then clarified by the addition of CH_3OH. White crystals then formed, which were collected in four crops to yield 5.32 g (65%) of **5** (mp 264–272 °C): mp 275–277 °C (CH_3OH); ir (Nujol) 3200 (NH), 1730 (ester C=O), 1670 cm^{-1} (C=O); NMR (Me_2SO-d_6) δ 8.20–7.08 (m, 6, aromatic), 3.72 (s, 3, CO_2CH_3); mass spectrum (70 eV) *m/e* 364 (molecular ion).

Anal. Calcd for $C_{16}H_{10}Cl_2N_2O_4$: C, 52.63; H, 2.76; N, 7.67. Found: C, 52.54; H, 2.70; N, 7.57.

B. From 4. A 1.66-g (4.98 mmol) quantity of **4** and 0.746 g (5.00 mmol) of 3-methyl-1-*p*-tolyltriazine (TMT, Eastman) in 50 ml of tetrahydrofuran (THF) were heated at reflux for 14 h. The solution was filtered to remove a small amount of insoluble material and the filtrate was concentrated to dryness. The resulting solid was slurried with ether and the white solid was collected to yield 1.13 g (62%) of **5**, mp 271–275 °C , whose ir (Nujol) was identical with that prepared in part A.

4-[[5-Chloro-2-(6-chloro-1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl)phenyl]carbonyl]morpholine (6). A 7.50-g (22.5 mmol) quantity of **3** was mixed with 75 ml of morpholine and heated at reflux for 15 min. The brown solution was cooled and diluted with water to produce a precipitate which was collected and air dried to yield 8.48 g (90%) of crude **6**: mp 244–246 °C (EtOH–H_2O); ir (Nujol) 1725, 1670 cm^{-1}; NMR (Me_2SO-d_6) δ 8.13–7.17 (m, 6, aromatic), 3.58 (broad signal, 8, morpholino); mass spectrum (70 eV) *m/e* 419 (molecular ion)

Anal. Calcd for $C_{19}H_{15}Cl_2N_3O_4$: C, 54.30; H, 3.60; N, 10.00. Found: C, 54.39; H, 3.56; N, 9.94.

Reaction of Isatoic Anhydride (1a) with 1-Isocyanatobenzoyl Chloride (2a). To a solution of 16.3 g (0.100 mol) of **1a** in 300 ml of dry dioxane was added a solution of 18.2 g (0.100 mol) of **2a** in 60 ml of dioxane and 1 ml of pyridine. After 6 days, an ir of a concentrated aliquot indicated the absence of the isocyanate stretching band of **2a** at 2280 cm^{-1}. The solution was concentrated and the gummy material was lixiviated with ether. The gum was then triturated with acetone and the resulting white solid was collected and air dried to yield 8.80 g (33%) of 5*H*,12*H*-quinazolino[3,2-*a*][3,1]benzoxazine-5,12-dione (**8**), mp 224–226 °C: mp 228–229 °C (acetone) (lit.[3] mp 228 °C); ir (Nujol) 1775, 1710, 1695 cm^{-1}; mass spectrum (70 eV) *m/e* 264 (molecular ion).

Anal. Calcd for $C_{15}H_8N_2O_3$: C, 68.18; H, 3.05; N, 10.60. Found: C, 68.50; H, 3.14; N, 10.45.

The filtrate was concentrated to an oily material which crystallized and was collected and washed with acetone to yield 4.60 g (13%) of 2-[1,4-dihydro-2,4-dioxo-3(2*H*)-quinazolinyl]benzoic acid (**7**), mp 280–284 °C: mp 295–296 °C (C_2H_5OH) (lit.[14] mp 298–300 °C); ir (Nujol) 1710, 1660 cm^{-1} (broad).

Anal. Calcd for $C_{15}H_{10}N_2O_4$: C, 63.83; H, 3.57; N, 9.93. Found: C, 63.80; H, 3.56; N, 9.84.

Preparation of 10-Chloro-5H,12H-quinazolino[3,2-a][3,1]-benzoxazine-5,12-dione (12). A 19.8-g (0.100 mol) quantity of **1b** and 18.2 g (0.100 mol) of **2a** were mixed with 400 ml of dioxane and 1 ml of pyridine and heated at reflux. After 3 days solution had resulted and after 10 days, the ir of an aliquot indicated the absence of **2a.** The solution was evaporated to a small volume and the white solid was collected to give 17.9 g (60%) of **12,** mp 243–247 °C: ir (Nujol) 1700, 1770 cm^{-1}; mass spectrum (70 eV) *m/e* 298 (molecular ion).

Anal. Calcd for $C_{15}H_7ClN_2O_3$: C, 60.32; H, 2.36; N, 9.37. Found: C, 60.20; H, 2.44; N, 9.40.

Preparation of 5-Chloro-2-[1,4-dihydro-2,4-dioxo-3(2H)-quinazolinyl]benzoic Acid (16a). Following a literature procedure for the preparation of anthraniloylanthranilic acid,[15] isatoic anhydride (**1a**) and 5-chloroanthranilic acid (**13a**) were condensed to give 2-[(aminobenzoyl)amino]-5-chlorobenzoic acid (**14a**): mp 227–229 °C (ethyl acetate); ir 3350–2300 (broad stretching, with spikes at 3330 and 3230), 1650 cm^{-1} (C=O); NMR (Me_2SO-d_6) δ 12.12 (broad s, 1, NH), 8.67 (d, J = 9 Hz, 1, H ortho to CONH), 8.03 (d, J = 3 Hz, 1, H ortho to CO_2H), 7.86–6.57 (m, 5, remaining aromatic).

Anal. Calcd for $C_{14}H_{11}ClN_2O_3$: C, 57.84; H, 3.81; N, 9.63. Found: C, 58.10; H, 3.90, N, 9.46.

A 28.8-g (0.0991) mol) quantity of **14a** and 15.5 g (0.104 mol) of TMT in 200 ml of THF were stirred at 25 °C for 24 h. The solution was concentrated, slurried with ether–hexane, and the product collected to yield 22.1 g (73%) of methyl 2-[(aminobenzoyl)amino]-5-chlorobenzoate (**15a**): mp 157–158 °C (C_2H_5OH); ir (Nujol) 3460, 3340, and 3230 (NH and NH_2), 1685 (ester C=O), 1645 cm^{-1} (amide C=O); NMR ($CDCl_3$) δ 11.77 (broad s, 1, NH), 8.85 (d, J = 9 Hz, 1, H ortho to CONH), 7.90 (d, J = 3 Hz, 1, H ortho to CO_2CH_3), 7.70–7.00 (m, 3, aromatic), 6.83–6.48 (m, 2, aromatic), 5.68 (broad s, 2, NH_2, D_2O exchangeable), 3.90 (s, 3, CH_3).

Anal. Calcd for $C_{15}H_{13}ClN_2O_3$: C, 59.11; H, 4.29; N, 9.19. Found: C, 59.40; H, 4.37; N, 9.43.

To 10.0 g (32.8 mmol) of **15a** in 100 ml of pyridine was added 3.56 g (32.8 mmol) of ethyl chloroformate and the solution was heated at reflux for 15 h. The solution was concentrated and partitioned between water and CH_2Cl_2 and the insoluble material was collected and air dried to yield 5.30 g (51%) of **16a:** mp 303–304.5 °C dec; ir (Nujol) 3250–2100 (NH and OH), 1715 (acid C=O), 1680 (amide C=O), 1650 cm^{-1} (urea C=O); NMR (Me_2SO-d_6) δ 11.63 (s, 1, CO_2H, D_2O exchangeable), 8.30–7.08 (m, 8, aromatic and NH).

Anal. Calcd for $C_{15}H_9ClN_2O_4$: C, 56.88; H, 2.86; N, 8.84. Found: C, 56.70; H, 3.06; N, 9.01.

Preparation of 2-[1,4-Dihydro-2,4-dioxo-6-chloro-3(2H)-quinazolinyl]benzoic Acid (16b). From 5-Chloroisatoic Anhydride (1b) and Anthranilic Acid (13b). Following a literature procedure for the preparation of anthraniloylanthranilic acid,[15] **1b** and **13b** were condensed to give 2-[(amino-5-chlorobenzoyl)amino]benzoic acid (**14b**) in 76% yield: mp 248–249 °C (C_2H_5OH); ir (Nujol) 3600–2300 (broad stretching, with spikes at 3420 and 3320), 1650 cm^{-1} (C=O).

Anal. Calcd for $C_{14}H_{11}ClN_2O_3$: C, 57.84; H, 3.81; N, 9.63. Found: C, 57.60; H, 3.87; N, 9.70.

Using the procedure described for the preparation of **15a, 14b** was converted to methyl 2-[(2-amino-5-chlorobenzoyl)amino]benzoate (**15b**) in 88% yield: mp 158–159 °C (C_2H_5OH); ir (Nujol) 3460, 3350, and 3300 (NH and NH_2), 1680 (ester C=O), 1660 cm^{-1} (amide C=O); NMR (Me_2SO-d_6) δ 11.20 (s, 1, NH), 8.38 (d, J = 8 Hz, 1, H ortho to CO_2CH_3), 8.10–6.67 (m, 6, remaining aromatic), 6.64 (broad s, 2, NH_2), 3.90 (s, 3, CH_3).

Anal. Calcd for $C_{15}H_{13}ClN_2O_3$: C, 59.11; H, 4.29; N, 9.19. Found: C, 59.00; H, 4.28; N, 9.37.

Using the procedure described for the preparation of **16a, 15b** was converted to **16b** (35%): mp 303 °C; ir (Nujol) 3250–2100 (NH and OH), 1720 (acid C=O), 1675 (amide C=O), 1650 cm^{-1} (urea C=O); NMR (Me_2SO-d_6) δ 11.80 (s, 1, CO_2H, D_2O exchangeable), 8.27–7.14 (m, 8, aromatic and NH).

Anal. Calcd for $C_{15}H_9ClN_2O_4$: C, 56.89; H, 2.86; N, 8.85. Found: C, 56.60; H, 2.94; N, 8.76.

B. From 12. A 3.60-g (12.0 mmol) quantity of **12** was slurried with 60 ml of CH_3OH and warmed on a steam bath. A solution of 25% $NaOCH_3$ in CH_3OH was added dropwise until solution resulted. Cooling produced white prisms which were collected to afford 82% of methyl 2-(6-chloro-1,4-dihydro-2,4-dioxo-3-quinazolinyl)benzoate (**17**): mp 269–272 °C; ir (Nujol) 3130 (NH), 1715 (ester C=O), 1650 cm^{-1} (C=O), NMR (Me_2SO-d_6) δ 8.14–7.70 (m, 2, aromatic protons ortho to C=O groups), 7.70–7.04 (m, 5, remaining aromatic), 3.69 (s, 3, CH_3).

Anal. Calcd for $C_{16}H_{11}ClN_2O_4$: C, 58.11; H, 3.35; N, 8.47. Found: C, 57.90; H, 3.44; N, 8.55.

A 2.00-g (6.05 mmol) quantity of **17** was slurried with 25 ml of 4 N NaOH and warmed gently until solution resulted. The solution was acidified with concentrated HCl and the resulting white solid was collected and air dried to yield 1.90 g (99%) of **16b,** mp 301–303 °C dec, which was spectrally identical with the material made in part A. A mixture melting point of this material and that made in part A was undepressed. A mixture melting point of **16b** and **16a** was substantially depressed (275–278 °C dec).

Acknowledgment. The authors wish to thank Barbara Isenbarger (Toxicology Department) for obtaining the spectral data.

Registry No.—**1a,** 118-48-9; **1b,** 4743-17-3; **2a,** 5100-23-2; **3,** 59187-49-4; **4,** 59187-50-7; **5,** 59187-51-8; **6,** 59187-52-9; **7,** 1701-95-7; **8,** 13969-02-3; **12,** 59187-53-0; **13a,** 635-21-2; **13b,** 118-92-3; **14a,** 40082-89-1; **14b,** 59187-54-1; **15a,** 59187-55-2; **15b,** 59187-56-3; **16a,** 59187-57-4; **16b,** 59187-58-5; **17,** 59187-59-6; morpholine, 110-91-8.

References and Notes

(1) N. P. Peet and S. Sunder, *J. Org. Chem.*, **40,** 1909 (1975).
(2) Y. Iwakura, K. Uno, and S. Kang, *J. Org. Chem.*, **31,** 142 (1966).
(3) G. Doleschall and K. Lempert, *Acta Chim. Acad. Sci. Hung.*, **48,** 77 (1966).
(4) Thionyl chloride in pyridine or phosphorus oxychloride in pyridine also effect the transformation of **7** to **8.**[3]
(5) (a) Compound **10** was first synthesized by K. Butler and M. W. Partridge, *J. Chem. Soc.*, 1512 (1959), and has more recently been prepared by others;[5b,c,d] (b) S. K. P. Sinha, *J. Indian Chem. Soc.*, **48,** 989 (1971); (c) M. Takahashi, S. Onizawa, and R. Shioda, *Nippon Kagaku Kaishi,* **8,** 1259 (1972); *Chem. Abstr.*, **78,** 72078q (1973); (d) S. Palazzo, L. I. Giannola, and M. Neri, *J. Heterocycl. Chem.*, **12,** 1077 (1975).
(6) A reviewer has suggested that the cycloaddition reaction which produces **11** in path a of Scheme VI is unlikely from a stereochemical standpoint.
(7) The direct conversion of **15a** and **15b** to **16a** and **16b,** respectively, was unexpected. We have previously reported[8] that compound **15,** where R = R′ = H, when treated with ethyl chloroformate in pyridine yields methyl 2-[[2-(ethoxycarbonylamino)benzoyl]amino]benzoate. The presence of the chlorine substituents in **15a** and **15b** has a profound effect on the reactions which produce **16a** and **16b,** respectively.
(8) N. P. Peet and S. Sunder, *J. Org. Chem.*, **39,** 1931 (1974).
(9) In path b of Scheme VI, the loss of HCl prior to the loss of CO_2 is as easily envisioned.
(10) R. P. Staiger, C. L. Moyer, and G. R. Pitcher, *J. Chem. Eng. Data,* **8,** 454 (1963). We thank a reviewer for calling this article to our attention.
(11) This quinazolinedione could also arise by nucleophilic attack of isatoic anhydride on phenyl isocyanate.
(12) Melting points are uncorrected. Ir spectra were recorded with a Perkin-Elmer 727B instrument; NMR spectra with Varian T-60 and Perkin-Elmer R32 (90 MHz) spectrometers; mass spectra with a Hitachi RMU-6D mass spectrometer. Combustion analyses were performed by Dow Analytical Laboratories.
(13) The NMR spectrum of **3** in Me_2SO-d_6 was identical with that of **4** taken in Me_2SO-d_6, indicating a rapid **3** → **4** conversion effected by water in the Me_2SO-d_6.
(14) G. Doleschall and K. Lempert, *Monatsh. Chem.*, **95,** 1083 (1964).
(15) R. P. Staiger and E. B. Miller, *J. Org. Chem.*, **24,** 1214 (1959).