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1. *Triazaphenanthrenes. Part III.*[1] *Synthesis of Some 9-Aryl-2 : 3 : 10-triazaphenanthrenes.*

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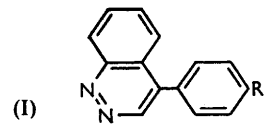
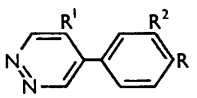
Cyclodehydration of the phenylpyridazines (II; R′ = NH·COAr) to 2 : 3 : 10-triazaphenanthrenes (III) is best effected with phosphorus pentoxide in nitrobenzene. The presence of a nitro-substituent prevents cyclisation in some instances. Methiodides of the triazaphenanthrenes were biologically inactive.

Of the twenty-eight possible triazaphenanthrene systems, derivatives of only five have been described. 1 : 3 : 9-Triazaphenanthrenes have been prepared[2,3] and the 1 : 2 : 9-system was described in Part II.[1] Two isolated 1 : 5 : 9- and 1 : 5 : 10-triazaphenanthrenes have been obtained as by-products in the synthesis of 2′ : 3-diamino-2 : 3′-dipyridyl from the corresponding diamide.[4] The synthesis of some 2 : 3 : 10-triazaphenanthrenes from 4-arylcinnolines (I) is now described.


(I)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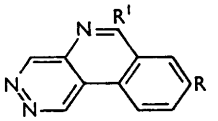
(III)

One of the key intermediates, 4-phenylcinnoline (I; R = H), was prepared[5] from *o*-aminobenzophenone which was available more readily on a large scale from *o*-benzoylbenzamide[6] than from toluene-*p*-sulphonylanthranilic acid.[7] Oxidation of the cinnoline (I; R = H) to 5-phenylpyridazine-3 : 4-dicarboxylic acid and subsequent decarboxylation in 2-ethoxyethanol provided a cleaner sample of the monoacid (II; R = R^2 = H, R^1 = CO_2H), m. p. 220° (decomp.), than that obtained by the literature method;[5] Stoermer and Fincke were in error in assigning this melting point to the dicarboxylic acid. Attempts to convert the monoacid into the amide (II; R = R^2 = H, R^1 = $CO{\cdot}NH_2$) *via* the acid chloride were unsuccessful, possibly because of cyclisation to the corresponding diazafluorenone.[8] However, the methyl ester gave the amide readily, from which the amine (II; R = R^2 = H, R^1 = NH_2) was obtained by treatment with potassium hypobromite. An attempt to prepare this amine directly from the monoacid by using hydroxylamine hydrochloride in polyphosphoric acid[9] failed.

[1] Part II, Atkinson and Mattocks, *J.*, 1957, 3722.
[2] Galland and Robinson, *J.*, 1925, **127**, 1493.
[3] Atkinson and Mattocks, *J.*, 1957, 3718.
[4] Brydowna, *Roczniki Chem.*, 1934, **14**, 304.
[5] Stoermer and Fincke, *Ber.*, 1909, **42**, 3115.
[6] Hewett, Lermit, Openshaw, Todd, Williams, and Woodward, *J.*, 1948, 292.
[7] Scheifile and De Tar, *Org. Synth.*, **32**, 8.
[8] Rule and Bretscher, *J.*, 1927, 925; Bretscher, Rule, and Spence, *J.*, 1928, 1493; Bell, *J.*, 1928, 3247.
[9] Snyder, *J. Amer. Chem. Soc.*, 1953, **75**, 2014.

Cyclodehydration of the benzamido-derivative (II; R = R^2 = H, R^1 = NHBz) was not readily achieved; thus, heating under reflux with phosphorus oxychloride alone[10] or mixed with nitrobenzene,[11] or with the further addition of stannic chloride [12] had no effect. The use of phosphorus pentoxide in boiling toluene or tetralin [13,14] gave similar results, but in nitrobenzene 9-phenyl-2 : 3 : 10-triazaphenanthrene (III; R = H, R^1 = Ph) was formed in yields of about 50%. Phosphorus pentoxide alone [14] gave no useful product, and although admixture with polyphosphoric acid [15] provided some (20%) of the triazaphenanthrene, difficulties in isolation discouraged variations of this method. An optimum yield of about 26% was obtained by cyclisation in a melt of aluminium chloride–sodium chloride,[16] so this, too, was discarded in favour of phosphorus pentoxide in nitrobenzene.

4-Amino-5-phenylpyridazine (II; R = R^2 = H, R^1 = NH_2) was readily converted into the *o*-, *m*-, and *p*-nitrobenzoyl derivatives, the last two of which were cyclised to the corresponding triazaphenanthrenes. The *o*-nitrobenzamido-derivative resisted cyclisation, and comparison with the preceding results suggests that steric factors are responsible; this is supported by Walls' failure [17] to prepare 7-nitro-9-*o*-nitrophenylphenanthridine and by his isolation of only a small yield of the 3-nitro-analogue.

Quaternisation of 9-*p*-nitrophenyl-2 : 3 : 10-triazaphenanthrene (III; R = H, R^1 = *p*-NO_2·C_6H_4) with dimethyl sulphate gave a mixture from which one pure monomethiodide was isolated. Reduction before quaternisation yielded a 9-*p*-aminophenyl-2 : 3 : 10-triazaphenanthrene monomethiodide, but in the *m*-aminophenyl series, no pure salt was isolated from treatment with methyl iodide in nitromethane. Reaction of 9-phenyl-2 : 3 : 10-triazaphenanthrene (III; R = H, R^1 = Ph) with methyl iodide yielded a monomethiodide, but attempts to form a diquaternary derivative failed.

A similar scheme from 4-*p*-methoxyphenylcinnoline (I; R = OMe) [18] was planned, but a derived triazaphenanthrene could not be synthesised. The methoxy-cinnoline (I; R = OMe) was oxidised to 5-*p*-methoxyphenylpyridazine-3 : 4-dicarboxylic acid as described by Stoermer and Gaus [18] who attributed to this acid the melting point which we found for the monocarboxylic acid (II; R = OMe, R^1 = CO_2H, R^2 = H). The diacid was best decarboxylated in 2-ethoxyethanol, the literature method giving only unchanged material; dry heating or decarboxylation in diethylene glycol gave a lower yield of less pure material. Nitration of the monoacid gave the *Bz*-nitro-derivative (II; R = OMe, R^1 = CO_2H, R^2 = NO_2), represented thus in view of the stability towards nitration of other pyridazine acids lacking the phenyl substituent [18] and the directive properties of the other groups present. Its amide was converted into the amine (II; R = OMe, R^1 = NH_2, R^2 = NO_2), a direct method from the acid and hydroxylamine [9] being unsuccessful. The benzamido-compound (II; R = OMe, R^1 = NHBz, R^2 = NO_2) could not be cyclised; this behaviour was not unexpected in view of the known deactivating effect of a nitro-group, especially *ortho* or *para* to the point at which ring-closure might occur.[10b] Replacement of the nitro-group by the ethoxycarbonylamino-group would probably overcome this,[19] but because of difficulties encountered in the reduction of the nitro-group only the compound (II; R = OMe, R^1 = NHBz, R^2 = NH_2) has been prepared.

Pure 9-*p*-aminophenyl-2 : 3 : 10-triazaphenanthrene monomethiodide and crude 9-*p*-nitro- and 9-*m*-nitro-phenyl-2 : 3 : 10-triazaphenanthrene methiodide are ineffective against

[10] (*a*) Morgan and Walls, *J.*, 1931, 2447; Walls, *J.*, 1934, 104; (*b*) Ritchie, *J. Proc. Roy. Soc. N.S.W.*, 1945, **78**, 147.
[11] Walls, *J. Soc. Chem. Ind.*, 1947, **66**, 182; *J.*, 1945, 294.
[12] Barber, Bretherick, Eldridge, Holt, and Wragg, *J. Soc. Chem. Ind.*, 1950, **69**, 82; Petrow and Wragg, *J.*, 1950, 3516; Nunn, Schofield, and Theobald, *J.*, 1952, 2797.
[13] Bischler and Napieralski, *Ber.*, 1893, **26**, 1903; Ritchie, *J. Proc. Roy. Soc. N.S.W.*, 1945, **78**, 134.
[14] Petrow, Stack, and Wragg, *J.*, 1943, 316.
[15] Taylor and Kalenda, *J. Amer. Chem. Soc.*, 1954, **76**, 1699; Badger and Sasse, *J.*, 1957, 4.
[16] Ebel, G.P. 614,196/1935: *Chem. Abs.*, 1935, **29**, 5859.
[17] Walls, *J.*, 1932, 2225.
[18] Stoermer and Gaus, *Ber.*, 1912, **45**, 3104.
[19] Walls, *J.*, 1947, 67.

P. berghei, *B. rodhaini*, *T. congolense*, *T. equiperdum*, and *T. cruzi* in mice; they were active against hæmolytic *Streptococcus*, *Staph. aureus*, *B. coli*, and *Candida albicans* only at concentrations of about 50 mg. % when tested in Hedley Wright broth.

Experimental

5-Phenylpyridazine-3 : 4-dicarboxylic Acid.—Hot aqueous potassium permanganate (200 g. in 2 l.) was added dropwise (*ca.* 3 hr.) with stirring to a suspension of 4-phenylcinnoline (40 g.) in water (3 l.) at *ca.* 90° and heating and stirring were then continued for a further hour. The excess of permanganate was destroyed by alcohol, and the manganese dioxide digested with hot water (*ca.* 300 c.c.). The filtrate and washings were combined and concentrated to *ca.* 800 c.c., cooled, and acidified with a large excess of dilute sulphuric acid to yield a pale yellow solid (44·4 g.), m. p. 148—150° (efferv.) [remelting at 214° (decomp.)].

5-Phenylpyridazine-4-carboxylic Acid.—The crude foregoing acid (40 g.) and ethyl 2-hydroxyethyl ether (120 c.c.) were heated to boiling in an oil-bath during *ca.* 15 min. Heating was continued until evolution of gas had almost ceased (3—4 min.), then the mixture was removed from the bath, allowed to cool, and filtered; the residue was washed with a small volume of alcohol and the crude mono-acid twice crystallised from methanol (charcoal), giving cubes of 5-phenylpyridazine-4-carboxylic acid (16—18 g., 46—51% based on 4-phenylcinnoline), m. p. 222—224° (decomp.) (Stoermer and Fincke [5] give m. p. 220—221°, decomp.) (Found: C, 66·2; H, 4·1; N, 15·0. Calc. for $C_{11}H_8O_2N_2$: C, 66·0; H, 4·0; N, 14·0%).

Methyl 5-Phenylpyridazine-4-carboxylate.—A solution of diazomethane in dioxan (*ca.* 400 c.c.; from 40 g. of *N*-methylnitrosourea) was added dropwise with stirring during 45 min. to a suspension of the powdered carboxylic acid [40 g.; m. p. 220—222° (decomp.)] in dioxan (1·5 l.) and stirring was continued until dissolution was complete (*ca.* 15 min.). Next day the clarified solution was concentrated under reduced pressure to *ca.* 120 c.c. on the steam-bath and poured into ice and water (*ca.* 1·2 l.). The crude *ester* on recrystallisation (charcoal) from light petroleum (b. p. 90—110°) formed needles (34·4 g., 80%), m. p. 103—105° (Found: C, 66·9; H, 4·8; N, 13·3. $C_{12}H_{10}O_2N_2$ requires C, 67·3; H, 4·7; N, 13·1%).

4-Carbamoyl-5-phenylpyridazine.—Powdered methyl 5-phenylpyridazine-4-carboxylate (20 g.) and methanolic ammonia (*ca.* 1 l.) [prepared by saturating methanol (1 l.) with dry ammonia (*ca.* 59 g.) in an ice bath] were kept at room temperature for 8 days. Removal of the solvent and storage in a vacuum desiccator gave the crude amide, which was purified by concentrating a chloroform (*ca.* 800 c.c.) solution; drying at 100° gave dull blades of *4-carbamoyl-5-phenylpyridazine* (16·3 g., 88%), m. p. 170—172° (Found: C, 66·2; H, 4·6; N, 21·3. $C_{11}H_9ON_3$ requires C, 66·3; H, 4·55; N, 21·1%).

4-Amino-5-phenylpyridazine.—Powdered 4-carbamoyl-5-phenylpyridazine (26 g.) was added in one portion to aqueous potassium hypobromite [370 c.c., from bromine (8·0 c.c.), potassium hydroxide (40 g.), and water (400 c.c.) at 0°] and stirred at 0° for 45 min. To this solution was added more aqueous potassium hydroxide (220 c.c., 10%), and the mixture was heated at 80° for 35 min. Cooling and crystallisation of the crude amine from water (charcoal) yielded long blades (18·0 g., 80%), m. p. 153—155°. Recrystallisation from benzene gave the pure *amine* as thick pale yellow blades, m. p. 154—156° (Found: C, 70·0; H, 5·2; N, 24·8. $C_{10}H_9N_3$ requires C, 70·15; H, 5·3; N, 24·55%).

4-Benzamido-5-phenylpyridazine.—4-Amino-5-phenylpyridazine was benzoylated with benzoyl chloride in pyridine on the steam bath for 3 hr. *4-Benzamido-5-phenylpyridazine* (88%) formed plates (from alcohol), m. p. 202—204° (Found: C, 73·7; H, 4·6; N, 15·65. $C_{17}H_{13}ON_3$ requires C, 74·2; H, 4·8; N, 15·3%).

9-Phenyl-2 : 3 : 10-triazaphenanthrene.—Phosphorus pentoxide (*ca.* 4·8 g.) was added to a solution of 4-benzamido-5-phenylpyridazine (2·4 g.) in dry nitrobenzene (48 c.c.) at *ca.* 130° and the mixture stirred at *ca.* 180° for 6 hr., with a further addition of phosphorus pentoxide (*ca.* 2·4 g.) after 3 hr. After cooling, water was added and nitrobenzene removed in steam. The aqueous residue was filtered hot and the filtrate made alkaline with dilute sodium hydroxide solution. The precipitate (1·7 g.) crystallised from aqueous acetone (charcoal) as dark yellow blades (1·2 g., 53%), m. p. 193—195°. Pure *9-phenyl-2 : 3 : 10-triazaphenanthrene* had m. p. 196—198° (from methanol) (Found: C, 79·2; H, 4·3; N, 16·4. $C_{17}H_{11}N_3$ requires C, 79·4; H, 4·3; N, 16·3%).

Quaternisation of 9-*Phenyl*-2 : 3 : 10-*triazaphenanthrene*.—(*a*) The base (1·2 g.), methyl iodide (4·8 c.c.), and methanol (24 c.c.) were heated under reflux for 3 hr. The solid was washed successively with methanol and ether and crystallised from water (*ca.* 140 c.c.), giving orange-yellow blades of a *monomethiodide* (1·5 g.), m. p. 285—287° (decomp.) (Found: C, 53·9; H, 3·4; N, 10·3; I, 32·4. $C_{18}H_{24}N_3I$ requires C, 54·1; H, 3·5; N, 10·5; I, 31·8%).

(*b*) The base (0·3 g.), dimethyl sulphate (0·3 c.c.), and nitrobenzene (30 c.c.) were heated at 160° (oil-bath) for 3 hr. The solution was diluted with ether, and the product extracted with water. The extract was saturated with potassium iodide, and the precipitate crystallised from water, forming orange-yellow microblades (270 mg.), m. p. 285—287° (decomp.), alone and on admixture with a sample prepared as above.

4-*Nitrobenzamido*-5-*phenylpyridazines*.—4-Amino-5-phenylpyridazine (2·5 g.) and *o*-nitrobenzoyl chloride (from 3·0 g. of the acid) in dry pyridine (20 c.c.) were heated on the steam-bath for 2½ hr. The product [from methanol (charcoal)] formed plates of 4-o-*nitrobenzamido*-5-*phenylpyridazine* (2·7 g., 58%), m. p. 216—218° (Found: C, 63·4; H, 3·4; N, 17·9. $C_{17}H_{12}O_3N_4$ requires C, 63·7; H, 3·8; N, 17·5%).

Prepared similarly, 4-m-*nitrobenzamido*-5-*phenylpyridazine* formed prisms (7·8 g., 83%; from 5·0 g.), m. p. 198—199°, from acetone (Found: C, 63·7; H, 3·9; N, 17·5%), and the 4-p-*nitrobenzamido*-compound, blades (5·9 g., 79%; from 4·0 g.), m. p. 216—217°, from methanol (Found: C, 63·5; H, 4·0; N, 18·0%).

9-p-*Nitrophenyl*-2 : 3 : 10-*triazaphenanthrene*.—Phosphorus pentoxide (*ca.* 18 g.) was added in three portions to a stirred solution of 5-*p*-nitrobenzamido-4-phenylpyridazine (6 g.) in dry nitrobenzene (180 c.c.) at *ca.* 140°, and the mixture was stirred at *ca.* 180° for 6 hr., with further addition of phosphorus pentoxide (*ca.* 9 g.) after 3 hr. Water was carefully added and the mixture was basified with 6N-sodium hydroxide, steam distilled, and filtered hot. The brown material was crystallised from nitromethane (charcoal), yielding fine needles of 9-p-*nitrophenyl*-2 : 3 : 10-*triazaphenanthrene* (3·9 g., 69%), m. p. 300—301° (Found: C, 67·3; H, 3·4; N, 18·9. $C_{17}H_{10}O_2N_4$ requires C, 67·5; H, 3·3; N, 18·6%).

9-p-*Aminophenyl*-2 : 3 : 10-*triazaphenanthrene*.—A warm solution of stannous chloride (10 g.) in concentrated hydrochloric acid (10 c.c.) and the nitro-compound (2 g.) were heated on the steam-bath for ½ hr. The mixture was then poured into 6N-sodium hydroxide, the solution heated almost to boiling, and the yellow precipitate crystallised from alcohol, furnishing golden-yellow needles of 9-p-*aminophenyl*-2 : 3 : 10-*triazaphenanthrene* (1·6 g., 89%), m. p. 305—307° (Found: C, 74·7; H, 4·3; N, 19·65. $C_{17}H_{12}N_4$ requires C, 75·0; H, 4·4; N, 20·6%).

Quaternisation of 9-p-*Aminophenyl*-2 : 3 : 10-*triazaphenanthrene*.—The base (2 g.) and methyl iodide (10 c.c.) in nitromethane (400 c.c.) were heated under reflux for 4 hr. The crude salt (2·12 g.), m. p. 320—323° (decomp.), was crystallised from a large volume of alcohol, yielding fine red needles of the *monomethiodide*, m. p. 323—324° (decomp.) (Found: C, 51·2; H, 3·8; N, 13·3; I, 31·2. $C_{18}H_{15}N_4I$ requires C, 52·05; H, 3·7; N, 13·5; I, 30·65%).

9-m-*Nitrophenyl*-2 : 3 : 10-*triazaphenanthrene*.—Prepared as described for the *p*-nitrophenyl compound 9-m-*nitrophenyl*-2 : 3 : 10-*triazaphenanthrene* crystallised from acetonitrile (charcoal) in fluffy needles (0·7 g., 74%; from 1 g.), m. p. 251—254° raised by a further crystallisation to 255—257° (Found: C, 67·5; H, 3·2; N, 17·8%).

9-m-*Aminophenyl*-2 : 3 : 10-*triazaphenanthrene*.—The nitro-compound (2·5 g.) was reduced as described for the *p*-isomer. Crystallisation from alcohol yielded yellow blades of 9-m-*aminophenyl*-2 : 3 : 10-*triazaphenanthrene* (1·8 g., 80%), m. p. 245—246° (Found: C, 74·15; H, 4·65; N, 21·0%).

5-p-*Methoxyphenylpyridazine*-4-*carboxylic Acid*.—The corresponding dicarboxylic acid [20 g., m. p. 148—150° (efferv.); prepared by oxidation of 4-*p*-methoxyphenylcinnoline] was heated with 2-ethoxyethanol (40 c.c.) under reflux until evolution of gas almost ceased (15 min.). The crude product was washed with a little alcohol. Crystallisation from alcohol (charcoal) gave needles of the *acid* (11 g., 62% based upon 4-*p*-methoxyphenylcinnoline), m. p. 206—208° (decomp.) (lit.,[18] m. p. 205°).

5-(4-*Methoxy*-3-*nitrophenyl*)*pyridazine*-4-*carboxylic Acid*.—A solution of 5-*p*-methoxyphenylpyridazine-4-carboxylic acid (37 g.) in dilute nitric acid (1000 c.c.) was evaporated almost to dryness on the steam-bath; the yellow solid was triturated with cold water (*ca.* 1 l.), filtered, and washed thoroughly with water. This crude material was dissolved in boiling dilute nitric acid and water added to give fine pale yellow needles of 5-(4-methoxy-3-nitrophenyl)pyridazine-4-carboxylic acid (31 g.), m. p. 232—234° (decomp.). Lit.,[18] m. p. 230° (decomp.).

The *methyl ester* (prepared by use of diazomethane in dioxan) formed yellow blades (78%), m. p. 151—153°, from alcohol (Found: C, 54·1; H, 4·0; N, 14·9. $C_{13}H_{11}O_5N_3$ requires C, 54·0; H, 3·8; N, 14·5%).

4-*Carbamoyl*-5-(4-*methoxy*-3-*nitrophenyl*)*pyridazine*.—A suspension of powdered methyl ester (30 g.) in methanol (1·5 l.) was saturated at 5° with ammonia and set aside at room temperature with occasional shaking for 14 days, being resaturated with ammonia after 7 days. The precipitate and material recovered from the filtrate by concentration and precipitation was crystallised from alcohol, yielding pale yellow blades of the *amide* (20·5 g., 72%), m. p. 233—234° (decomp.) (Found: C, 52·4; H, 3·8; N, 21·2. $C_{12}H_{10}O_4N_4$ requires C, 52·55; H, 3·7; N, 20·4%).

4-*Amino*-5-(4-*methoxy*-3-*nitrophenyl*)*pyridazine*.—Powdered 4-carbamoyl-5-(4-methoxy-3-nitrophenyl)pyridazine (10 g.) was added in one portion to aqueous potassium hypobromite [from bromine (2·0 c.c.) and potassium hydroxide (10 g.) in water (100 c.c.) at 0°] and the mixture stirred at 0° for 7 hr. The solution was heated on the steam-bath for 40 min. and the crude amine then crystallised from acetone, giving needles of 4-*amino*-5-(4-*methoxy*-3-*nitrophenyl*)*pyridazine* (4·5 g., 50%), m. p. 199—201° (decomp.) (Found: C, 53·9; H, 4·4; N, 22·35. $C_{11}H_{10}O_3N_4$ requires C, 53·7; H, 4·1; N, 22·8%).

4-*Benzamido*-5-(4-*methoxy*-3-*nitrophenyl*)*pyridazine*.—The foregoing amine was benzoylated by benzoyl chloride–pyridine in boiling chlorobenzene for 32 hr. Golden needles (9·9 g.), m. p. 206—210°, separated from the cold solution, and recrystallisation from absolute ethanol (charcoal) gave the *benzamide* as pale yellow needles (62%), m. p. 220—221° (Found: C, 61·8; H, 3·8; N, 15·3. $C_{18}H_{14}O_4N_4$ requires C, 61·7; H, 4·0; N, 16·0%).

4-(3-*Amino*-4-*methoxyphenyl*)-5-*benzamidopyridazine*.—(*a*) The foregoing nitro-compound (0·6 g.) was heated in ethanol (60 c.c.) at *ca.* 90° with hydrogen under pressure (*ca.* 100 atm.) for 2½ hr., in the presence of 5% palladised charcoal. The solution was filtered and evaporated to dryness, and the residue crystallised (twice) from methanol (charcoal), yielding needles of the *amine* (0·27 g.), m. p. 185—186° (Found: C, 67·4; H, 5·2; N, 17·1. $C_{18}H_{16}O_2N_4$ requires C, 67·5; H, 5·0; N, 17·5%).

(*b*) The powdered nitro-compound (4 g.) was added portionwise to stannous chloride reagent [20] (150 c.c.) and set aside at room temperature. Next day, the white solid was treated with ice and an excess of dilute ammonia solution. A chloroform extract was washed, dried ($MgSO_4$), and evaporated (finally with the aid of benzene under reduced pressure). The residue crystallised from methanol (charcoal) as pale yellow needles (2·7 g., 74%), m. p. 185—186° alone and on admixture with a sample prepared as above.

4-(3-*Acetamido*-4-*methoxyphenyl*)-5-*benzamidopyridazine*.—The foregoing amine was acetylated with acetic anhydride on the steam bath for ½ hr. 4-(3-*Acetamido*-4-*methoxyphenyl*)-5-*benzamidopyridazine* (0·3 g., 66%) formed flat needles, m. p. 189—190°, from acetone (charcoal) (Found: C, 66·1; H, 4·9; N, 15·2. $C_{20}H_{18}O_3N_4$ requires C, 66·3; H, 5·0; N, 15·5%).

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[20] Albert and Linnell, *J.*, 1936, 1617.