67. *Nitro- and Amino-3-arylphthalaz-4-ones, and Corresponding 1-Methyl Compounds.*

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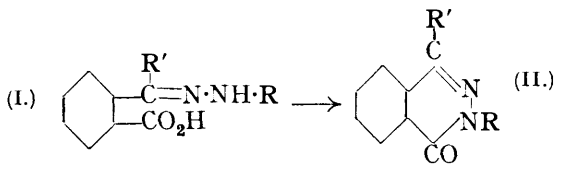
As a preliminary to other projected work, it was necessary to prepare and characterise as reference compounds the series of nitro- and amino-3-arylphthalaz-4-ones, and the corresponding 1-methyl compounds, now described. The opportunity was taken to study also the stabilities of the intermediate hydrazones (I, or its lactone form) and their relative ease of conversion into the nitro-3-arylphthalaz-4-ones (II) under standard conditions.



(R = nitroaryl; R′ = H or Me)

o-Phthalaldehydic acid was condensed with nitroarylhydrazines, and ring closure of the resulting *o*-carboxybenzaldehydenitroarylhydrazones (I; R′ = H) to the nitro-3-arylphthalaz-4-ones (II; R′ = H) effected in various ways (cf. Racine, *Annalen*, 1887, **239**, 86; Rowe and co-workers, J., 1928, 2551, 2557; 1932, 475). Similarly, acetophenone-*o*-carboxylic acid gave nitro-3-aryl-1-methylphthalaz-4-ones (II; R′ = Me) *via* the *o*-carboxyacetophenonenitroarylhydrazones (I; R′ = Me) (cf. Rowe and co-workers, J., 1931, 1920; 1935, 1135).

The stabilities of the intermediate hydrazones (I; R′ = H or Me) vary considerably with the position of substituents in the 3-phenyl nucleus. Thus, the comparative stabilities are in the order R = 2′-nitrophenyl>2′-halogeno- and 2′ : 6′-dihalogeno-4′-nitrophenyl> 4′-nitro-2′-methylphenyl>4′-nitrophenyl>3′-nitrophenyl. Similar retarding effects of *o*-substituents in a phenyl side-chain on formation of six-membered rings were observed with phthalylnitroarylhydrazides (J., 1935, 1810) and with sodium benzaldehydenitroaryl-

hydrazone-ω-sulphonate-2-β-acrylic acids (J., 1935, 1796). In general, the hydrazones (I; R′ = H) are more stable than the corresponding hydrazones (I; R′ = Me), *e.g.*, some conditions which converted the latter into the nitro-3-aryl-1-methylphthalaz-4-ones in excellent yields caused no, or only partial, ring closure with most of the former examined. A simple method of preparing large quantities of (II; R′ = H or Me), applicable in most cases with good results, is to dissolve (I; R′ = H or Me) in cold concentrated sulphuric acid and leave the solution over-night, but the behaviour of (I; R′ = H, R = 2′ : 6′-dibromo-4′-nitrophenyl) was exceptional, as it was unaltered by this treatment.

The nitro-compounds (II; R′ = H or Me) are more stable than the corresponding 1-ones and, unlike the latter, neither form salts with mineral acids nor dissolve in aqueous alkalis, although they dissolve slowly on boiling with aqueous-alcoholic sodium hydroxide and then give a highly coloured layer on addition of acetone.

Amino-3-aryl-1-methylphthalaz-4-ones are obtained by reducing (II; R′ = Me) with aqueous-alcoholic sodium sulphide, but amino-3-arylphthalaz-4-ones in most cases are best prepared from (I; R′ = H) with aqueous sodium sulphide, which effects both ring closure and reduction.

Experimental.

o-*Carboxybenzaldehydenitroarylhydrazones* (I; R′ = H).—These compounds (or their lactone forms) were readily prepared by refluxing a mixture of alcoholic solutions of equimolecular proportions of *o*-phthalaldehydic acid and the appropriate nitroarylhydrazine (cf. J., 1928, 2555).

Methods of converting (I) *into Nitro-3-arylphthalaz-4-ones* (II; R′ = H).—In all cases 1 g. of (I) was used. (i) Heating at 20° below the m. p. of the corresponding compound (II) for 5 minutes; refluxing with (ii) alcohol (20 c.c.) for 1 hour, or (iii) glacial acetic acid (10 c.c.) for ¼ hour, or (iv) nitrobenzene (10 c.c.) for ¼ hour, or (v) amyl alcohol (10 c.c.), saturated with dry hydrogen chloride, for 1 hour, or (vi) acetic anhydride (5 c.c.) for 10 minutes, or (vii) acetic anhydride (10 c.c.) and pyridine (1 c.c.) for 3 hours; (viii) dissolution in cold concentrated sulphuric acid (10 c.c.), the solution being left over-night. Any unchanged (I) was extracted with cold aqueous sodium carbonate solution, or with cold sodium hydroxide solution when (I) was not very soluble in the former.

Amino-3-arylphthalaz-4-ones (II; R′ = H, R = aminoaryl).—Except where R = 3′-aminophenyl (see below), these compounds were best made from (I), rather than from (II; R = nitroaryl). A suspension of (I) (10 g.) in water (250 c.c.) was heated with sodium sulphide crystals (80 g.) in water (100 c.c.) at 50—60° until the bluish-red colour of the solution changed to orange-yellow or yellow and almost colourless crystals had separated.

o-*Carboxybenzaldehyde-2′-nitrophenylhydrazone* (I; or its lactone form) crystallised from alcohol in bright orange needles, m. p. 229° (refluxed for ¼ hour; yield, 85%) (Found: N, 14·8. $C_{14}H_{11}O_4N_3$ requires N, 14·7%), soluble in cold sodium carbonate or hydroxide solution with a violet colour. Conversion into (II; R = 2′-nitrophenyl): Methods (i) to (v) nil; (vi) 90%; (vii) and (viii) 95%.

2′-*Nitro-3-phenylphthalaz-4-one* (II) crystallised from benzene or glacial acetic acid in colourless prisms, m. p. 201° (Found: C, 63·25; H, 3·7; N, 16·0. $C_{14}H_9O_3N_3$ requires C, 62·9; H, 3·4; N, 15·7%), insoluble in aqueous alkalis and hydrochloric acid. This compound and its analogues described subsequently were best prepared by method (viii), unless otherwise stated.

2′-*Amino-3-phenylphthalaz-4-one* crystallised from aqueous alcohol in colourless needles, m.p. 184° (yield, 4 g.; 48·1%) (Found: C, 70·8; H, 4·4; N, 17·8. $C_{14}H_{11}ON_3$ requires C, 70·9; H, 4·6; N, 17·7%); the *acetyl* derivative crystallised from aqueous alcohol in colourless needles, m. p. 237° (Found: N, 15·3. $C_{16}H_{13}O_2N_3$ requires N, 15·05%).

I; R = 3′-*Nitrophenyl* (cf. J., 1928, 2563) (refluxed for 5 minutes; yield, 71%). Conversion into (II; R = 3′-nitrophenyl): Method (i) complete; (ii) 16%; (iii) to (viii) complete.

II; R = 3′-*Aminophenyl*.—In this case, treatment of (I) with aqueous sodium sulphide merely gave (II; R = 3′-nitrophenyl) in a form which was not reduced by prolonged boiling. Reduction was best effected by refluxing a fine suspension of (II; R = 3′-nitrophenyl) (4 g.) in water (100 c.c.) and alcohol (100 c.c.) with sodium sulphide crystals (30 g.) for 20 minutes (yield, 2·3 g.; 64·8%). The *acetyl* derivative of 3′-amino-3-phenylphthalaz-4-one (*loc. cit.*) crystallised from aqueous alcohol in colourless plates, m. p. 225° (Found: N, 15·2%).

I; R = 4′-*Nitrophenyl* (cf. J., 1928, 2555) (refluxed for 5 minutes; yield, 92%). Conversion into (II; R = 4′-nitrophenyl): Method (i) complete, (ii) 5%; (iii) to (viii) complete.

II; R = 4′-*Aminophenyl* (*loc. cit.*) (yield, 4·1 g.; 49·3%).

I; R = 4′-*Nitro-2′-methylphenyl* (cf. J., 1932, 482) (refluxed for ½ hour; yield, 87%). Conversion into (II; R = 4′-nitro-2′-methylphenyl): Methods (i) and (ii) nil; (iii) 57%; (iv) 31%; (v) 85%; (vi) 71%; (vii) 95%; (viii) 83%.

II; R = 4′-*Amino-2′-methylphenyl* crystallised from aqueous alcohol in colourless needles, m. p. 187—188° (yield, 7·5 g.; 89·3%) (Found: N, 16·8. $C_{15}H_{13}ON_3$ requires N, 16·7%); the *acetyl* derivative crystallised from aqueous alcohol in colourless needles, m. p. 214° (Found: N, 14·5. $C_{17}H_{15}O_2N_3$ requires N, 14·3%).

I; R = 2′-*Chloro-4′-nitrophenyl* crystallised from alcohol in small yellow needles, m. p. 229° (refluxed for 10 minutes; yield, 75%) (Found: N, 13·3. $C_{14}H_{10}O_4N_3Cl$ requires N, 13·1%), soluble in sodium carbonate and hydroxide solutions with orange-red and deep bluish-red colours, respectively. Conversion into (II; R = 2′-chloro-4′-nitrophenyl): Methods (i) to (v) nil; (vi) 32%; (vii) 99%; (viii) 70%.

II; R = 2′-*Chloro-4′-nitrophenyl*, best prepared by method (vi) but refluxing for 3 hours, crystallised from glacial acetic acid or alcohol in colourless prisms, m. p. 171° (Found: N, 13·9. $C_{14}H_8O_3N_3Cl$ requires N, 13·9%), insoluble in cold aqueous alkalis, but soluble in hot aqueous-alcoholic sodium hydroxide with a bluish-red colour.

II; R = 2′-*Chloro-4′-aminophenyl* crystallised from aqueous alcohol in colourless needles, m. p. 186° (yield, 6·1 g.; 71·8%) (Found: N, 15·6. $C_{14}H_{10}ON_3Cl$ requires N, 15·45%); the *acetyl* derivative crystallised from aqueous alcohol in colourless needles, m. p. 247° (Found: N, 13·6. $C_{16}H_{12}O_2N_3Cl$ requires N, 13·4%).

I; R = 2′ : 6′-*Dichloro-4′-nitrophenyl* crystallised from alcohol in orange-yellow needles, m. p. 225—226° (refluxed for 10 minutes; yield, 85%) (Found: N, 11·7. $C_{14}H_9O_4N_3Cl_2$ requires N, 11·85%), soluble in sodium carbonate and hydroxide solutions with orange-red and intense reddish-violet colours, respectively. Conversion into (II; R = 2′: 6′-dichloro-4′-nitrophenyl): Methods (i) to (v) nil; (vi) 70%; (vii) 93%; (viii) 82%.

II; R = 2′ : 6′-*Dichloro-4′-nitrophenyl*, prepared in the same way as the 2′-chloro-analogue, crystallised from glacial acetic acid in small colourless prisms, m. p. 179° (Found: Cl, 21·1. $C_{14}H_7O_3N_3Cl_2$ requires Cl, 21·1%), soluble in boiling aqueous-alcoholic sodium hydroxide with a deep red colour.

II; R = 2′ : 6′-*Dichloro-4′-aminophenyl* crystallised from aqueous alcohol in colourless prisms, m. p. 226° (yield, 3·8 g.; 44%) (Found: N, 13·8; Cl, 23·2. $C_{14}H_9ON_3Cl_2$ requires N, 13·7; Cl, 23·2%); the *acetyl* derivative crystallised from aqueous alcohol in colourless needles, m. p. 281° (Found: N, 12·1. $C_{16}H_{11}O_2N_3Cl_2$ requires N, 12·05%).

I; R = 2′-*Bromo-4′-nitrophenyl* crystallised from alcohol in yellow needles, m. p. 204° (refluxed for ½ hour; yield, 86%) (Found: N, 11·7. $C_{14}H_{10}O_4N_3Br$ requires N, 11·5%), soluble in sodium carbonate and hydroxide solutions with orange-red and reddish-violet colours, respectively. Conversion into (II; R = 2′-bromo-4′-nitrophenyl): Methods (i) to (iii) nil; (iv) 31%; (v) 23%; (vi) 94%; (vii) and (viii) 96%.

II; R = 2′-*Bromo-4′-nitrophenyl* crystallised from glacial acetic acid in colourless prisms, m. p. 154° (Found: C, 48·3; H, 2·5; Br, 23·4. $C_{14}H_8O_3N_3Br$ requires C, 48·55; H, 2·3; Br, 23·1%), soluble in boiling aqueous-alcoholic sodium hydroxide with a bluish-red colour.

II; R = 2′-*Bromo-4′-aminophenyl* crystallised from aqueous alcohol in colourless prisms, m. p. 203° (yield, 5·3 g.; 61·1%) (Found: C, 53·4; H, 3·4. $C_{14}H_{10}ON_3Br$ requires C, 53·2; H, 3·2%); the *acetyl* derivative crystallised from aqueous alcohol in small colourless prisms, m. p. 251° (Found: N, 12·0. $C_{16}H_{12}O_2N_3Br$ requires N, 11·7%).

I; R = 2′ : 6′-*Dibromo-4′-nitrophenyl* crystallised from alcohol in brownish-yellow needles, m. p. 220° (refluxed for 10 minutes; yield, 86%) (Found: Br, 36·0. $C_{14}H_9O_4N_3Br_2$ requires Br, 36·1%), soluble in sodium carbonate and hydroxide solutions with yellow and deep bluish-violet colours, respectively. Conversion into (II; R = 2′ : 6′-dibromo-4′-nitrophenyl): Methods (i) and (ii) nil; (iii) 36%; (iv) 40%; (v) 92%; (vi) 81%; (vii) 96%; (viii) nil.

II; R = 2′ : 6′-*Dibromo-4′-nitrophenyl*, best prepared by method (vi) but refluxing for 2 hours, crystallised from alcohol in colourless prisms, m. p. 190° (Found: N, 9·8; Br, 37·9. $C_{14}H_7O_3N_3Br_2$ requires N, 9·9; Br, 37·65%).

II; R = 2′ : 6′-*Dibromo-4′-aminophenyl* crystallised from aqueous alcohol in colourless prisms, m. p. 255° (yield, 5 g.; 56·1%) (Found: N, 10·8; Br, 40·5. $C_{14}H_9ON_3Br_2$ requires N, 10·6; Br, 40·5%); the *acetyl* derivative crystallised from aqueous alcohol in small colourless prisms, m, p. 257° (Found: N, 9·6. $C_{16}H_{11}O_2N_3Br_2$ requires N, 9·6%).

o-*Carboxyacetophenonenitroarylhydrazones* (I; R′ = Me).—These compounds (or their lactone forms) were prepared by adding a solution of the appropriate nitroarylhydrazine in alcohol

(sufficient to prevent precipitation of the unaltered hydrazine) to an aqueous solution of an equimolecular proportion of acetophenone-*o*-carboxylic acid at about 70°, unless otherwise stated (cf. J., 1931, 1923).

Conversion of (I) *into Nitro-3-aryl-1-methylphthalaz-4-ones* (II; R′ = Me).—Methods (i) to (viii) as described for the unmethylated compounds (p. 312) were used.

Amino-3-aryl-1-methylphthalaz-4-ones (II; R′ = Me, R = aminoaryl).—These were prepared satisfactorily in all cases from finely divided (II; R = nitroaryl).

o-*Carboxyacetophenone-o-nitrophenylhydrazone* (I; or its lactone form) (*loc. cit.*). Conversion into (II; R = 2′-nitrophenyl): Method (i) nil; (ii) 6%; (iii) 60%; (iv) to (viii) almost complete.

II; R = 2′-*Aminophenyl*.—(II; R = 2′-nitrophenyl) (5 g.), sodium sulphide crystals (50 g.), water (125 c.c.), and alcohol (125 c.c.) were refluxed for 1 hour. 2′-*Amino-3-phenyl-1-methylphthalaz-4-one* crystallised from alcohol in pale yellow needles, m. p. 239° (yield, 3·2 g.; 71·6%) (Found: N, 16·4. $C_{15}H_{13}ON_3$ requires N, 16·7%); the *acetyl* derivative crystallised from alcohol in colourless needles, m. p. 241° (Found: N, 13·95. $C_{17}H_{15}O_2N_3$ requires N, 14·3%).

I; R = 3′-*Nitrophenyl*, when the condensation was carried out at 15—20°, separated as a bright orange, amorphous powder, which was very unstable and did not melt, but became colourless at 115°, owing to formation of (II; R = 3′-nitrophenyl) (*loc. cit.*), and then melted at 167° (yield, 82%) (Found: N, 14·0. $C_{15}H_{13}O_4N_3$ requires N, 14·05%); it was soluble in cold sodium carbonate and hydroxide solutions with orange colours. Conversion into (II; R = 3′-nitrophenyl): Method (i) complete; (ii) 78%; (iii) to (viii) complete.

II; R = 3′-*Aminophenyl*, prepared in the same way as the 2′-isomeride, but with sodium sulphide crystals (25 g.) and refluxing for 3 hours, crystallised from alcohol in almost colourless prisms, m. p. 173° (yield, 2·5 g.; 56%) (Found: N, 16·9%); the *acetyl* derivative crystallised from alcohol in colourless prisms, m. p. 220° (Found: N, 14·5%).

I; R = 4′-*Nitrophenyl* (*loc. cit.*). Conversion into (II; R = 4′-nitrophenyl): Method (i) complete; (ii) 71%; (iii) 85%; (iv) to (viii) complete.

II; R = 4′-*Aminophenyl*, best prepared from (I; R = 4′-nitrophenyl) (5 g.), sodium sulphide crystals (40 g.), and water (40 c.c.), heated at 50° for 3 hours, crystallised from alcohol in almost colourless, stout prisms, m. p. 206—207° (yield, 2·8 g.; 66·7%) (Found: C, 71·3; H, 5·4; N, 16·9. $C_{15}H_{13}ON_3$ requires C, 71·7; H, 5·2; N, 16·7%); the *acetyl* derivative crystallised from alcohol in colourless needles, m. p. 252° (Found: N, 14·0%).

I; R = 4′-*Nitro-2′-methylphenyl*, when the condensation was carried out at 5—10°, separated as a bright yellow powder, m. p. 145° (Found: N, 13·3. $C_{16}H_{15}O_4N_3$ requires N, 13·4%), soluble in cold sodium carbonate and hydroxide solutions with orange and deep red colours, respectively. Conversion into (II; R = 4′-nitro-2′-methylphenyl): Method (i) nil; (ii) 29%; (iii) 72%; (iv) to (viii) complete.

II; R = 4′-*Nitro-2′-methylphenyl* crystallised from glacial acetic acid in colourless prisms, m. p. 178° (Found: C, 65·4; H, 4·5; N, 14·2. $C_{16}H_{13}O_3N_3$ requires C, 65·1; H, 4·4; N, 14·2%), sparingly soluble in boiling aqueous-alcoholic sodium hydroxide with a red colour, converted into bluish-red on addition of acetone.

II; R = 4′-*Amino-2′-methylphenyl*, prepared in the same way as the 2′-amino-analogue, but with sodium sulphide crystals (25 g.), water (140 c.c.), and alcohol (140 c.c.), and refluxing for 2 hours, crystallised from alcohol in very pale yellow prisms, m. p. 191° (yield, 2·6 g.; 57·9%) (Found: N, 15·95. $C_{16}H_{15}ON_3$ requires N, 15·85%); the *acetyl* derivative crystallised from alcohol in small colourless prisms, m. p. 235° (Found: N, 14·0. $C_{18}H_{17}O_2N_3$ requires N, 13·7%).

I; R = 2′-*Chloro-4′-nitrophenyl* crystallised from alcohol in orange-yellow needles, m. p. 160° (Found: N, 12·9. $C_{15}H_{12}O_4N_3Cl$ requires N, 12·6%), soluble in sodium carbonate and hydroxide solutions with a red colour. Conversion into (II; R = 2′-chloro-4′-nitrophenyl): Method (i) nil; (ii) 4%; (iii) to (viii) complete.

II; R = 2′-*Chloro-4′-nitrophenyl* crystallised from alcohol in small colourless prisms, m. p. 206° (Found: C, 57·0; H, 3·3; N, 13·1. $C_{15}H_{10}O_3N_3Cl$ requires C, 57·05; H, 3·2; N, 13·3%), sparingly soluble in aqueous-alcoholic sodium hydroxide with a cherry-red colour.

II; R = 2′-*Chloro-4′-aminophenyl*, prepared in the same way as the 2′-amino-analogue, but refluxing for 10 minutes, crystallised from alcohol in colourless prisms, m. p. 197° (yield, 2·3 g.; 50·8%) (Found: N, 14·7. $C_{15}H_{12}ON_3Cl$ requires N, 14·7%); the *acetyl* derivative crystallised from alcohol in small colourless prisms, m. p. 247° (Found: N, 12·6. $C_{17}H_{14}O_2N_3Cl$ requires N, 12·8%).

I; R = 2′ : 6′-*Dichloro-4′-nitrophenyl* crystallised from alcohol in small yellow needles, m. p. 135° (yield, 84%) (Found: N, 11·3. $C_{15}H_{11}O_4N_3Cl_2$ requires N, 11·4%). Conversion into (II; R = 2′ : 6′-dichloro-4′-nitrophenyl): Method (i) nil; (ii) 13%; (iii) to (viii) complete.

II; R = 2′ : 6′-*Dichloro*-4′-*nitrophenyl* crystallised from alcohol in small colourless needles or prisms, m. p. 235° (Found : C, 51·0; H, 2·6; Cl, 20·6. $C_{15}H_9O_3N_3Cl_2$ requires C, 51·4; H, 2·55; Cl, 20·3%).

II; R = 2′ : 6′-*Dichloro*-4′-*aminophenyl*, prepared in the same way as the 2′-amino-analogue, but with alcohol (250 c.c.), crystallised from alcohol in small colourless prisms, m. p. 279° (yield, 2·4 g.; 52·5%) (Found : Cl, 22·3. $C_{15}H_{11}ON_3Cl_2$ requires Cl, 22·2%); the *acetyl* derivative crystallised from glacial acetic acid in small colourless needles, m. p. 320° (Found : N, 11·4. $C_{17}H_{13}O_2N_3Cl_2$ requires N, 11·6%).

I; R = 2′-*Bromo*-4′-*nitrophenyl* (cf. J., 1935, 1137). Conversion into (II; R = 2′-bromo-4′-nitrophenyl) : Method (i) nil; (ii) 2%; (iii) to (viii) almost complete.

II; R = 2′-*Bromo*-4′-*aminophenyl*, prepared in the same way as the 2′-amino-analogue, but with sodium sulphide crystals (25 g.) and refluxing for ¼ hour, crystallised from alcohol in almost colourless needles, m. p. 130° (decomp.) after softening at 123° (yield, 2·8 g.; 61·1%) (Found : Br, 23·9. $C_{15}H_{12}ON_3Br$ requires Br, 24·2%); the *acetyl* derivative crystallised from acetic anhydride in small colourless prisms, m. p. 255° (Found : N, 11·3. $C_{17}H_{14}O_2N_3Br$ requires N, 11·3%).

I; R = 2′ : 6′-*Dibromo*-4′-*nitrophenyl* could not be obtained satisfactorily by the use of an aqueous solution of acetophenone-*o*-carboxylic acid. A boiling solution of acetophenone-*o*-carboxylic acid (20 g., colourless needles, m. p. 116°) in alcohol (100 c.c.) was added to a boiling solution of 2 : 6-dibromo-4-nitrophenylhydrazine (28 g.) (J., 1935, 1814) in alcohol (500 c.c.); on cooling, the *hydrazone* crystallised in small, pale yellow needles, m. p. 167° (yield, 35 g.) (Found : N, 9·3. $C_{15}H_{11}O_4N_3Br_2$ requires N, 9·2%). Conversion into (II; R = 2′ : 6′-dibromo-4′-nitrophenyl) : Method (i) nil; (ii) 18%; (iii) to (viii) almost complete.

II; R = 2′ : 6′-*Dibromo*-4′-*nitrophenyl* crystallised from glacial acetic acid in small colourless prisms, m. p. 237° (Found : C, 41·3; H, 2·3; Br, 36·2. $C_{15}H_9O_3N_3Br_2$ requires C, 41·0; H, 2·05; Br, 36·4%).

II; R = 2′ : 6′-*Dibromo*-4′-*aminophenyl*, prepared in the same way as the 2′-bromo-analogue, but with sodium sulphide crystals (40 g.), crystallised from alcohol in colourless needles, m. p. 274° (yield, 3 g.; 64·4%) (Found : C, 44·2; H, 2·9. $C_{15}H_{11}ON_3Br_2$ requires C, 44·0; H, 2·7%); the *acetyl* derivative crystallised from alcohol in small colourless needles, m. p. 315° (Found : N, 9·4. $C_{17}H_{13}O_2N_3Br_2$ requires N, 9·3%).

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