595 N-*Oxides of Some Hydroxy- and Amino-quinolines.*

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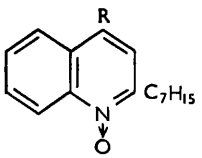
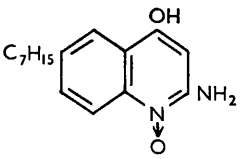
The *N*-oxides of certain hydroxy- and amino-quinolines bearing a heptyl substituent group have been prepared, but attempts to synthesise 2-amino-4-hydroxyquinoline 1-oxide were unsuccessful.

Lightbown [1] has isolated from *Pseudomonas pyocyanea* cultures a mixture which was an antagonist of dihydrostreptomycin and which, in higher concentrations, inhibited the growth of *Staphylococcus aureus*.[2] The mixture was shown by Cornforth and James [3] to contain 2-heptyl-4-hydroxyquinoline 1-oxide (I; R = OH) and smaller amounts of 2-octyl and 2-non-2′-enyl compounds. In contrast, Lott and Shaw [4] (see also refs. 5 and 6) reported that 4-hydroxyquinoline 1-oxide does not show antibacterial properties, although the 2-hydroxy-isomer is significantly active. In view of the properties thus conferred by the heptyl group, we prepared the compounds (I; R = OH and NH_2) and some related compounds for more general biological examination.

Although Cornforth and James [3] synthesised the oxide (I; R = OH), details have not yet been published.* Our first projected synthesis requires 2-heptyl-4-nitroquinoline 1-oxide which should be a convenient source of both the 4-amino- and the 4-hydroxy-compound (cf. Ochiai [7]). 2-Heptylquinoline 1-oxide could not, however, be nitrated, although Ishikawa [8] described the nitration of quinaldine *N*-oxide. As an alternative, 2-heptyl-4-hydroxyquinoline was converted into the 4-chloro- and thence into the 4-benzyloxy-compound. Oxidation of the latter with perbenzoic acid [4] afforded 4-benzyloxy-2-heptylquinoline 1-oxide which was catalytically debenzylated to give the desired oxide (I; R = OH) in 48% overall yield. Similarly 6-heptyl-4-hydroxyquinoline was converted into the corresponding oxide.



(I)



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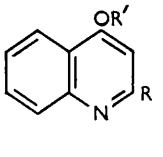

(III)

As a preliminary to the preparation of alkyl-substituted 4-aminoquinoline 1-oxides, the readily accessible 4-amino-7-chloroquinoline was used. Its diacetyl derivative with perbenzoic acid furnished the *N*-oxide which was hydrolysed to the amino-oxide; 4-amino-2- and 4-amino-6-heptylquinoline 1-oxide were prepared similarly. 2-Heptyl-4-*p*-tolylthioquinoline was obtained by condensation of toluene-*p*-thiol with 4-chloro-2-heptylquinoline but could not be oxidised to the sulphonyl-*N*-oxide. Finally, we hoped to prepare 2-amino-6-heptyl-4-hydroxyquinoline 1-oxide (II); preliminary experiments with 2-acetamido-4-acetoxyquinoline (III; R = NHAc, R′ = Ac) did not give the *N*-oxide. Then ethyl 4-chloroquinaldate was converted into 4-benzyloxyquinaldic acid and its methyl ester; the latter did not react with perbenzoic acid but gave the hydrazide from which 4-benzyloxy-2-diacetylaminoquinoline (III; R = NAc_2, R′ = CH_2Ph) was obtained by the Curtius reaction. Although this compound reacted readily with perbenzoic acid, no oxide could be isolated and much of the reactant was recovered. Side-reactions evidently occur to some extent during the oxidations with perbenzoic acid and

* (*Added, June 26th*, 1956.) Cornforth and James (*Biochem. J.*, 1956, **63**, 124) have recently described the synthesis of the oxide (I; R = OH) and two homologues by three methods.

[1] Lightbown, *J. Gen. Microbiol.*, 1954, **11**, iv.
[2] Lightbown and Jackson, *Biochem. J.*, 1954, 58, xlix.
[3] Cornforth and James, *ibid.*, p. xlviii.
[4] Lott and Shaw, *J. Amer. Chem. Soc.*, 1949, **71**, 70.
[5] Newbold and Spring, *J.*, 1948, 1864.
[6] Cunningham, Newbold, Spring, and Stark, *J.*, 1949, 2091.
[7] Ochiai, *J. Org. Chem.*, 1953, **18**, 534.
[8] Ishikawa, *J. Pharm. Soc. Japan*, 1945, **3**, A, 4.

probably account for the rather low yields in some of the other oxidations described; Lott and Shaw [2] also found that reaction of perbenzoic acid with some benzyloxy-compounds was unsatisfactory.

Jaffé [9] has recently shown (contrast earlier work [4,10]) that 4-hydroxy- and 4-aminopyridine 1-oxide exist in solution as such and not as the tautomeric 1-hydroxypyrid-4-one (and the corresponding imine). The compounds described above are therefore considered by analogy to have the *N*-oxide forms (Lott and Shaw [4] suggested 1-hydroxyquinol-4-one structures).

Experimental

Substituted Quinoline 1-*Oxides*.—The quinoline in chloroform (*ca.* 2 c.c./g.) was cooled at 0° while perbenzoic acid (1·0—1·1 mol.; 0·2—0·4M-solution in chloroform) was added gradually. After being kept at 0° overnight, the mixture was washed with sodium hydrogen carbonate solution and water, dried ($MgSO_4$), and evaporated under reduced pressure.

2-*Heptylquinoline*.—A stirred solution of sodamide (from 23 g. of sodium) in liquid ammonia was treated successively with quinaldine (143 g.) and hexyl bromide (165 g.) in ether (150 c.c.). The mixture was stirred under reflux for 2 hr. and then allowed to evaporate overnight. Isolated by addition of water (1 l.) and extraction with carbon tetrachloride, 2-*heptylquinoline* (53 g.) distilled as a colourless oil, b. p. 118—122°/0·2 mm., n_D^{20} 1·5499 (Found : C, 84·1; H, 9·6; N, 6·3. $C_{16}H_{21}N$ requires C, 84·5; H, 9·3; N, 6·2%). The *picrate* separated from methanol in rods, m. p. 101—102° (Found : C, 58·3; H, 5·4. $C_{22}H_{24}O_7N_4$ requires C, 57·9; H, 5·3%).

2-*Heptylquinoline* 1-*Oxide*.—The base (42 g.) was oxidised by the general procedure to give the *oxide* (24 g.) as a pale yellow oil, b. p. 172°/0·6 mm., which solidified and crystallised from light petroleum (b. p. 40—60°) in needles, m. p. 37—38° (Found : C, 78·5; H, 8·6; N, 5·9. $C_{16}H_{21}ON$ requires C, 79·0; H, 8·7; N, 5·8%).

4-*Benzyloxy*-2-*heptylquinoline*.—2-Heptyl-4-hydroxyquinoline (45 g.; Wells [11]) was refluxed with phosphorus oxychloride (125 c.c.) for 1 hr. The mixture was poured on ice and basified with 5N-sodium hydroxide, the product being isolated with ether. 4-*Chloro*-2-*heptylquinoline* (45 g.) had b. p. 124—127°/0·1 mm., n_D^{20} 1·5642 (Found : C, 73·3; H, 7·5; N, 5·4. $C_{16}H_{20}NCl$ requires C, 73·4; H, 7·7; N, 5·4%).

A solution of potassium hydroxide (13·2 g.) in benzyl alcohol (170 c.c.) and xylene (35 c.c.) was refluxed until no more water could be removed (phase separator). The chloro-amine (26·3 g.) was added and the mixture heated (bath 210°) for 2 hr. and then poured into water, the product being isolated with ether. 4-*Benzyloxy*-2-*heptylquinoline* (26·2 g.) was obtained as a colourless oil, b. p. 210—215°/0·5 mm., n_D^{20} 1·5815 (Found : C, 82·8; H, 8·0; N, 4·5. $C_{23}H_{27}ON$ requires C, 82·8; H, 8·2; N, 4·2%).

2-*Heptyl*-4-*hydroxyquinoline* 1-*Oxide*.—The foregoing benzyloxyquinoline (26·2 g.) was oxidised with perbenzoic acid by the general procedure. The crude solvent-free product was hydrogenated in ethanol (150 c.c.) over 5% palladised strontium carbonate (2 g.), and the solution filtered at the b. p. On cooling, the hydroxy-oxide (13·5 g.; m. p. 153—155°) separated; recrystallisation from ethyl methyl ketone gave needles, m. p. 156—157° (Found : C, 73·6; H, 8·0; N, 5·6. Calc. for $C_{16}H_{21}O_2N$: C, 74·1; H, 8·2; N, 5·4%).

Ethyl p-*Heptylanilinomethylenemalonate*.—A mixture of *p*-heptylaniline (48 g.), ethyl ethoxymethylenemalonate (55 g.), and benzene (200 c.c.) was refluxed and ethanol removed azeotropically during 30 min. Evaporation under reduced pressure afforded the *ester* (94 g.), n_D^{20} 1·5525 (Found : C, 69·4; H, 8·5; N, 4·0. $C_{21}H_{31}O_4N$ requires C, 69·8; H, 8·7; N, 3·9%).

3-*Ethoxycarbonyl*-6-*heptyl*-4-*hydroxyquinoline*.—Diphenyl–diphenyl ether (450 c.c.) was refluxed while the foregoing ester (46 g.) was added dropwise during 20 min. After the solution had been refluxed for a further 30 min., it was cooled and poured into light petroleum (b. p. 40—60°; 900 c.c.). Recrystallisation of the precipitate from ethanol furnished 3-*ethoxycarbonyl*-6-*heptyl*-4-*hydroxyquinoline* (50 g.), m. p. 233° (Found : C, 72·5; H, 8·0; N, 4·6. $C_{19}H_{25}O_3N$ requires C, 72·4; H, 8·0; N, 4·4%).

6-*Heptyl*-4-*hydroxyquinoline*.—The ester (50 g.) was hydrolysed for 30 min. with boiling 2N-sodium hydroxide (400 c.c.). Acidification gave 6-*heptyl*-4-*hydroxyquinoline*-3-*carboxylic acid* (23 g.) which separated from aqueous acetone in needles, m. p. 192—193° (Found : C, 71·3;

[9] Jaffé, *J. Amer. Chem. Soc.*, 1955, **77**, 4445, 4451.
[10] Shaw, *ibid.*, 1949, **71**, 67.
[11] Wells, *J. Biol. Chem.*, 1952, 196, 335.

H, 7·3; N, 4·8. $C_{17}H_{21}O_3N$ requires C, 71·1; H, 7·4; N, 4·9%). The acid (10 g.) was added during 10 min. to refluxing diphenyl–diphenyl ether (50 c.c.). The cooled solution was poured into light petroleum (b. p. 40—60°), and the precipitate recrystallised from acetone to give *6-heptyl-4-hydroxyquinoline* (8·4 g.) as prisms, m. p. 129—130° (Found: C, 79·3; H, 8·7; N, 5·9. $C_{16}H_{21}ON$ requires C, 79·0; H, 8·7; N, 5·8%).

6-Heptyl-4-hydroxyquinoline 1-Oxide.—Treatment of the hydroxyquinoline (8 g.) with phosphorus oxychloride as in the previous example gave *4-chloro-6-heptylquinoline* (7·5 g.), b. p. 137°/0·15 mm., f. p. 35° (Found: C, 73·4; H, 7·7; N, 5·3. $C_{16}H_{20}NCl$ requires C, 73·4; H, 7·7; N, 5·4%). The chloride was condensed with potassium benzyl oxide in the manner described and the product distilled. The fraction of b. p. 156—186°/0·1 mm. was oxidised with perbenzoic acid, according to the general procedure, and the crude product hydrogenated in ethanol (100 c.c.) in the presence of 10% palladised strontium carbonate (1 g.). Filtered at the b. p. and allowed to cool, the solution deposited the *hydroxy-oxide* (3·8 g.), needles, m. p. 183°, from ethanol (Found: C, 74·5; H, 8·2; N, 5·8%).

7-Chloro-4-diacetylaminoquinoline.—A solution of 4-amino-7-chloroquinoline (10 g.) in acetic anhydride (100 c.c.) and pyridine (50 c.c.) was refluxed for 1 hr. and then evaporated to dryness *in vacuo*. Repeated recrystallisation from benzene gave the solvated *diacetyl derivative*, prisms, m. p. 109—111° (Found: C, 63·5; H, 4·6; N, 9·6; Cl, 11·0. $C_{13}H_{11}O_2N_2Cl$ requires C, 63·7; H, 4·7; N, 9·3; Cl, 11·8%). The solvent of crystallisation could not be removed even at 80°/0·1 mm. Johnson, Woroch, and Buell [12] gave m. p. 197·2—198·2° but their compound was probably the monoacetyl derivative; only analytical data for carbon and hydrogen were reported and the values for mono- and di-acetyl derivatives are almost identical.

4-Amino-7-chloroquinoline 1-Oxide.—Oxidation of the diacetylamino-compound in the manner described (1·25 mol. of perbenzoic acid used) gave crude product, m. p. 135—137° (55%). *7-Chloro-4-diacetylaminoquinoline 1-oxide* separated from ethyl methyl ketone–light petroleum (b. p. 60—80°) in needles, m. p. 137—138° (Found: C, 56·3; H, 3·9; N, 10·1. $C_{13}H_{11}O_3N_2Cl$ requires C, 56·0; H, 4·0; N, 10·1%).

The oxide (7·6 g.) and 10% sodium hydroxide solution (75 c.c.) were boiled for 5 min.; the product separated on cooling. *4-Amino-7-chloroquinoline 1-oxide* (2·5 g.) crystallised from water in yellow needles, m. p. 246° (decomp.) (Found: C, 55·9; H, 3·7; N, 14·2. $C_9H_7ON_2Cl$ requires C, 55·5; H, 3·6; N, 14·4%).

4-Amino-2-heptylquinoline.—4-Chloro-2-heptylquinoline (45 g.) was added to phenol (450 g.), previously saturated at 80° with ammonia, and the mixture heated at 180° in a stream of ammonia for 5 hr. The solution was poured into 2N-sodium hydroxide, the product being isolated with ether. The *amine* (30 g.) formed rods, m. p. 101—103°, from ethyl methyl ketone–light petroleum (b. p. 60—80°) (Found: C, 78·9; H, 9·3; N, 11·6. $C_{16}H_{22}N_2$ requires C, 79·3; H, 9·2; N, 11·5%).

4-Diacetylamino-2-heptylquinoline.—The amino-quinoline (1 g.) was refluxed with acetic anhydride (10 c.c.) for 1·5 hr. Distillation afforded the *diacetyl derivative* (1·0 g.), yellow oil, b. p. 188—190°/0·4 mm., n_D^{20} 1·5589 (Found: C, 73·5; H, 7·9; N, 8·6. $C_{20}H_{26}O_2N_2$ requires C, 73·6; H, 8·0; N, 8·6%).

4-Amino-2-heptylquinoline 1-Oxide.—4-Amino-2-heptylquinoline (28 g.) was refluxed with acetic anhydride (300 c.c.) for 4 hr. The solution was evaporated *in vacuo* and the residue treated with perbenzoic acid according to the general procedure. The crude product was treated with 2·5N-sodium hydroxide (200 c.c.) and ethanol (100 c.c.); after the mixture had been refluxed for 30 min., ethanol was removed by distillation. On cooling, a sticky solid separated and this was recrystallised from ethyl methyl ketone to give the *amino-oxide* (12 g.), yellow rectangular plates, m. p. 176—177° (Found: C, 74·6; H, 8·6; N, 10·9. $C_{16}H_{22}ON_2$ requires C, 74·4; H, 8·6; N, 10·8%).

4-Amino-6-heptylquinoline.—This amine (3·5 g.) was obtained from the 4-chloro-compound (7 g.) as in the case of the isomer. It crystallised from light petroleum (b. p. 60—80°) in needles, m. p. 95° (Found: C, 79·3; H, 9·2; N, 11·6%). A solution of the amine (3·5 g.) in acetic anhydride (30 c.c.) and pyridine (15 c.c.) was refluxed for 45 min. and then evaporated under reduced pressure. Distillation afforded *4-diacetylamino-6-heptylquinoline* (2·7 g.) as a viscous, yellow oil, b. p. 166—168°/0·2 mm. (Found: C, 73·4; H, 8·0%).

4-Amino-6-heptylquinoline 1-Oxide.—Oxidation of the diacetyl derivative (2·7 g.) with perbenzoic acid (general procedure) gave the crude oxide which was refluxed for 1 hr. with acetic acid (20 c.c.) and 2N-sulphuric acid (10 c.c.). Basification with 6N-ammonia, followed

[12] Johnson, Woroch, and Buell, *J. Amer. Chem. Soc.*, 1949, **71**, 1901.

by recrystallisation from ethyl methyl ketone–ethanol, gave the yellow *oxide*, m. p. 211—212° (Found: C, 74·3; H, 8·4; N, 11·0%).

2-*Heptyl*-4-p-*tolythioquinoline*.—Sodium (0·5 g.) was dissolved in ethanol (25 c.c.) and toluene-*p*-thiol (3·3 g.) and 4-chloro-2-heptylquinoline (5 g.) were added, successively. The mixture was stirred under reflux (bath 100°) for 4 hr. and poured into water, the product being isolated with ether. Distillation gave the *sulphide* as a pale yellow oil, b. p. 197—198°/0·2 mm., n_D^{20} 1·6107 (Found: C, 78·8; H, 7·6; N, 3·5. $C_{23}H_{27}NS$ requires C, 79·1; H, 7·8; N, 4·8%).

2-*Acetamido*-4-*acetoxyquinoline*.—2-Amino-4-hydroxyquinoline (1·0 g.; Hardman and Partridge [13]) was refluxed with acetic anhydride (10 c.c.) for 30 min. The *product* (0·9 g.; m. p. 164—167°), which separated on cooling, formed plates, m. p. 168—169°, from benzene–light petroleum (b. p. 60—80°) (Found: C, 63·6; H, 4·9; N, 11·5. $C_{13}H_{12}O_3N_2$ requires C, 63·9; H, 5·0; N, 11·5%).

2-*Amino*-6-*heptyl*-4-*hydroxyquinoline*.—p-*Heptylanilinium toluene*-p-*sulphonate*, prepared in the usual manner, crystallised from aqueous methanol in needles, m. p. 120—121° (Found: C, 63·6; H, 8·0; N, 3·7. $C_{20}H_{29}O_3NS,H_2O$ requires C, 63·0; H, 8·2; N, 3·7%).

The salt (85 g.) and ethyl cyanoacetate (27 g.) were heated (bath 210°) for 1·3 hr. (cf. ref. 13). Trituration of the melt with ether gave 2-*amino*-6-*heptyl*-4-*hydroxyquinolinium toluene*-p-*sulphonate* (23 g.) which separated from ethyl methyl ketone–ethanol in needles, m. p. 229—230° (Found: C, 64·5; H, 6·9; N, 6·5. $C_{22}H_{30}O_4N_2S$ requires C, 64·2; H, 7·0; N, 6·5%). A solution of this salt (16 g.) in ethanol (50 c.c.) was treated with aqueous ammonia (20 c.c.) and then water (700 c.c.), to precipitate the product. 2-*Amino*-6-*heptyl*-4-*hydroxyquinoline* formed prisms, m. p. 276—277°, from ethyl methyl ketone–ethanol (Found: C, 73·7; H, 8·2. $C_{16}H_{22}ON_2$ requires C, 74·4; H, 8·6%).

2-*Ethoxycarbonyl*-6-*heptyl*-4-*hydroxyquinoline*.—*p*-Heptylaniline (19 g.) in acetic acid (50 c.c.) was gradually added to ethyl sodio-oxaloacetate (22 g.), the mixture being cooled below 10°. Next day, ice was added and the solution basified; the anil was isolated (ethyl acetate) and added dropwise during 20 min. to boiling diphenyl ether–diphenyl (300 c.c.). The solution was refluxed for 30 min. and then evaporated *in vacuo*. The *quinoline* (11·6 g.) crystallised from ethanol–ethyl acetate in rectangular plates, m. p. 152—153° (Found: C, 72·2; H, 7·6. $C_{19}H_{25}O_3N$ requires C, 72·3; H, 8·0%).

4-*Benzyloxyquinaldic Acid* (with W. A. Jones).—Ethyl 4-chloroquinaldate was prepared as described by Campaigne, Cline, and Kaslow.[14] The *amide*, prepared in aqueous-ethanolic ammonia, formed needles, m. p. 211—213°, from ethanol (Found: C, 58·1; H, 3·4; N, 13·6. $C_{10}H_7ON_2Cl$ requires C, 57·9; H, 3·4; N, 13·2%). The *hydrazide*, prepared in ethanol, crystallised from ethanol in needles, m. p. 176° (decomp.) (Found: C, 53·8; H, 3·5; Cl, 16·0. $C_{10}H_8ON_3Cl$ requires C, 54·2; H, 3·6; Cl, 16·0%).

To a solution of potassium benzyl oxide (prepared from potassium hydroxide, 7 g., as described) was added the chloro-ester (15 g.), and the mixture was refluxed for 1 hr. After addition of water (200 c.c.) the solution was distilled slowly for 1 hr. and, on cooling, the potassium salt crystallised. It was filtered off, washed with ether, and dissolved in hot water (200 c.c.); the solution was brought to about pH 6 with sulphuric acid and acetic acid to precipitate the benzyloxy-acid (8 g.). More product (2·5 g.) was obtained similarly from the original aqueous layer. Recrystallisation of the crude product [(m. p. 180—182° (decomp.)] from ethyl acetate and methanol gave the acid as fine needles, m. p. 185—187° (decomp.). Nakayama [15] gives m. p. 189° (decomp.) for a sample prepared from 4-benzyloxyquinoline 1-oxide.

The *methyl ester* (4 g.), prepared by reaction of the acid (5 g.) with diazomethane, separated from acetone–light petroleum (b. p. 60—80°) in needles, m. p. 130—132° (Found: C, 73·7; H, 5·1. $C_{18}H_{15}O_3N$ requires C, 73·7; H, 5·2%). The *amide* crystallised from ethanol in needles, m. p. 150—151° (Found: C, 73·4; H, 4·9; N, 10·0. $C_{17}H_{14}O_2N_2$ requires C, 73·4; H, 5·1; N, 10·1%).

2-*Acetamido*-4-*benzyloxyquinoline*.—The foregoing methyl ester (5·0 g.) in 2-methoxyethanol (25 c.c.) was refluxed with hydrazine hydrate (2 c.c.) for 5 hr. Concentration of the solution gave the *hydrazide* (3·6 g.), which formed rods, m. p. 146—147°, from 2-methoxyethanol (Found: C, 69·6; H, 5·1; N, 14·2. $C_{17}H_{15}O_2N_3$ requires C, 69·6; H, 5·2; N, 14·3%). This reaction did not proceed satisfactorily in ethanol.

Sodium nitrite (0·85 g.) in water (2·5 c.c.) was added to the hydrazide (3·6 g.) in benzene–acetic acid (40 c.c.; 1 : 1) at −5°, and the mixture was cooled below 0° for 30 min. After

[13] Hardman and Partridge, *J.*, 1954, 3880.
[14] Campaigne, Cline, and Kaslow, *J. Org. Chem.*, 1950, **15**, 600.
[15] Nakayama, *J. Pharm. Soc. Japan*, 1950, **70**, 423.

dilution with benzene (125 c.c.), the solution was gradually poured into ice-cold 1·5N-sodium carbonate (650 c.c.). The aqueous layer was extracted with benzene, and the combined organic layers were concentrated (to *ca.* 50 c.c.) under reduced pressure. After addition of acetic anhydride (50 c.c.), the mixture was distilled until the b. p. reached 95° and then refluxed for 45 min. Evaporation *in vacuo*, followed by recrystallisation from benzene–light petroleum (b. p. 60—80°), afforded 2-*acetamido*-4-*benzyloxyquinoline* (1·2 g.), needles, m. p. 197—199° (Found : C, 74·7; H, 5·2. $C_{18}H_{16}O_2N_2$ requires C, 74·0; H, 5·5%).

4-*Benzyloxy*-2-*diacetylaminoquinoline*.—When the Curtius reaction was repeated on a larger scale (14 g. of hydrazide), a mixture (8·2 g.) of the mono- and the di-acetyl derivative was obtained (Found : Ac, 19·8%). The sample was therefore refluxed with acetic anhydride (80 c.c.) for 3 hr. Isolated as above, the *diacetyl derivative* (4·5 g.) separated from benzene–light petroleum (b. p. 60—80°) in prisms, m. p. 146—147° (Found : C, 72·2; H, 5·5; N, 8·4; Ac, 26·0. $C_{20}H_{18}O_3N_2$ requires C, 71·8; H, 5·4; N, 8·4; Ac, 25·8%).

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