REDUCTION OF ACRIDINE AND ITS QUATERNARY SALTS BY HYDRAZINES

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In a continuation of our research on the aminoarylation of quaternary salts of acridine (I) [1], we studied the condensation of I with phenylhydrazine (II). Compound II was heated with N-methylacridinium iodide in dimethylformamide at 110°C for 30 min. Instead of the expected p-(9-acridanyl)phenylhydrazine (III) or other products that may result when III is oxidized by air oxygen [2], we isolated N,N'-dimethyl-diacridanyl (IV) in 73% yield.

The highest yield is obtained when the reaction is carried out in alcohol. Compound IV has an IR spectrum that is identical to the spectrum of N,N'-dimethyldiacridanyl synthesized by the Albert method [3] and does not depress the melting point of a sample obtained by the Albert method. Compound IV was also obtained by the reaction of II with N-methylacridinium chloride or N-methylacridinium methylsulfate in 69 and 90% yields, respectively. Phenylhydrazine reduces acridine to diacridanyl (V). The formation of diacridanyls in the reaction with II can be explained by the fact that the reductive properties of II predominate over the nucleophilic properties in this transformation. Compounds IV and V were also obtained by reduction of N-methylacridinium iodide and acridine by hydrazine hydrate.

EXPERIMENTAL

N,N'-Dimethyldiacridanyl (IV). A solution of 1.6 g (0.005 mole) of N-methylacridinium iodide and 1.08 g (0.01 mole) of phenylhydrazine [or 0.5 g (0.01 mole) of hydrazine hydrate] in 10 ml of alcohol was refluxed for 30 min. The precipitated IV was filtered to give 0.85 g (88%) [0.8 g (82%)] of a product with mp 280-281° (from dimethylformamide). Found: C 86.2; H 6.3; N 7.9%. $C_{28}H_{24}N_2$. Calculated: C 86.5; H 6.2; N 7.2%. IR spectrum in mineral oil, cm⁻¹: 1596, 1429, 1347, 1283, 1268, 1192, 1162, 1136, 1069, 1043, 934, 890, 760, 750, 638, 589, 558, and 490.

Diacridanyl (V). A solution of 1.79 g (0.01 mole) of acridine and 2.16 g (0.02 mole) of phenylhydrazine [or 1 g (0.02 mole) of hydrazine hydrate] in 6 ml of dimethylformamide was heated at 130° for 5 h. The precipitate was removed by filtration to give 1.3 g (72%) [0.25 g (14%)] of V with mp>300° (from dimethylformamide). Found: C 86.6; H 5.6; N 7.9%. $C_{26}H_{20}N_2$. Calculated: C 86.6; H 5.6; N 7.8%.

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