SYNTHESIS OF TETRAHYDROBENZIMIDAZOLE DERIVATIVES

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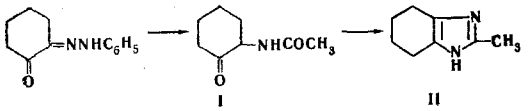
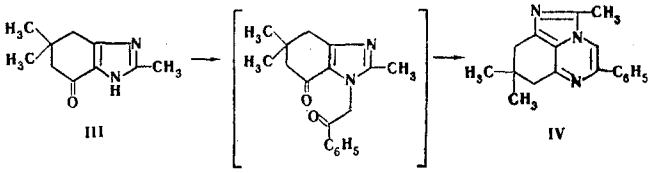
Continuing our study of monoarlyhydrazones of di- and tricarbonyl compounds with the aim of synthesizing various heterocycles [1], we proposed a new method for the preparation of tetrahydrobenzimidazoles. The reductive acylation of cyclohexane-1,2-dione monophenylhydrazone gave 2-acetamidocyclohexanone (I), which is converted to high yields of 2-methyltetrahydrobenzimidazole (II) by the action of ammonium acetate in acetic acid.



I II

3,5,5-Trimethyl-7-ketotetrahydrobenzimidazole (III) was similarly obtained from 5,5-dimethylcyclohexane-1,2,3-trione monophenylhydrazone. Treatment of the sodium derivative of III with bromoacetophenone and then with ammonium acetate in acetic acid gave the previously unknown 2,8,8-trimethyl-5-phenyl-7,8-dihydro-9H-pyrazino[3,2,1-h,i]benzimidazole (IV), which was isolated as the picrate.



III IV

EXPERIMENTAL

2-Acetamidocyclohexanone (I). This compound was obtained in 71% yield as colorless crystals with mp 95-96° (from ether). Found: C 62.0; H 8.5; N 9.0%. $C_8H_{13}NO_2$. Calculated: C 61.9; H 8.4; N 9.0%.

2-Methyl-4,5,6,7-tetrahydrobenzimidazole (II). This compound was obtained in 70.5% yield as white crystals with mp 220-221° (from alcohol) (mp 220-221° [2]).

2,5,5-Trimethyl-7-keto-4,5,6,7-tetrahydrobenzimidazole (III). This compound was obtained in 74% yield and had mp 113-114° (from benzene). Found: C 67.6; H 7.9; N 15.7%. $C_{10}H_{14}N_2O$. Calculated: C 67.4; H 7.9; N 15.7%.

Picrate of 2,8,8-Trimethyl-5-phenyl-9H-7,8-dihydropyrazino[3,2,1-h,i]benzimidazole (IV). This compound was obtained in 52% yield and had mp 260-265° (with sublimation). Found: C 57.0; H 4.3; N 16.6%. $C_{18}H_{19}N_3 \cdot C_6H_3N_3O_7$. Calculated: C 56.9; H 4.4; N 16.6%.

LITERATURE CITED

1. V. I. Shvedov, L. B. Altukhova, L. A. Chernyshkova, and A. N. Grinev, Khim.-Farmats. Zh., No. 10, 15 (1969).
2. R. Weidenhagen and H. Wegner, Ber., 71, 2124 (1938).

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