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REACTION OF 2-HYDRAZINO- AND 2-(BENZYLHYDRAZINO)BENZIMIDAZOLES WITH 4-PHENYL-3-BUTYN-2-ONE

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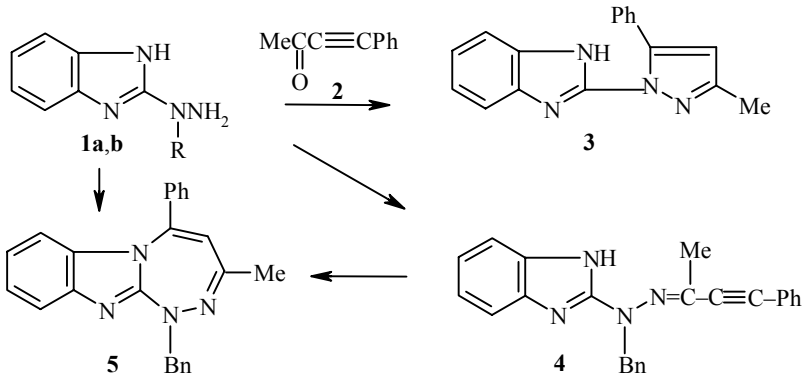
Keywords: 1-benzyl-3-methyl-5-phenyl-1,2,4-triazepino[4,3-*a*]benzimidazole, 2-hydrazinobenzimidazole, 2-(3-methyl-5-phenyl-1-pyrazolyl)benzimidazole, 4-phenyl-3-butyn-2-one, condensation.

In a study of the heterocyclization of hydrazinobenzimidazoles using dinucleophilic reagents [1-4], we were the first to investigate the reaction of 2-hydrazino- (**1a**) and 2-benzylhydrazinobenzimidazoles (**1b**) with 4-phenyl-3-butyn-2-one (**2**).

Heating equimolar amounts of **1a** and **2** in methanol at reflux with base catalysis for 3 h gave 2-(3-methyl-5-phenyl-1-pyrazolyl)benzimidazole (**3**).

The reaction of benzylhydrazinobenzimidazole **1b** with ketone **2** under the same conditions stops at the formation of the 1-(2-benzimidazolyl)-1-benzylhydrazone of 4-phenyl-3-butyn-2-one (**4**).

The reaction of 10 mmoles **1b** and 15 mmoles **2** in DMF at reflux for 4 h with two or three drops of triethylamine gave 1-benzyl-3-methyl-5-phenyl-1,2,4-triazepino[4,3-*a*]benzimidazole (**5**). Alternatively, **5** was obtained in 84% yield upon heating hydrazone **4** in DMF at reflux for 2 h.



1 a R = H, **b** R = CH_2Ph

Benzimidazole 3 was obtained in 71% yield; mp 250°C (methanol). IR spectrum (KBr), ν, cm^{-1}: ν_{NH} 3080. ^1H NMR spectrum (CD_3OD), δ, ppm: 2.68 (3H, s, CH_3); 6.64 (1H, s, CH); 7.19-7.88 (9H, m, H arom). Found, %: C 74.40; H 5.02; N 20.30. $C_{17}H_{14}N_4$. Calculated, %: C 74.43; H 5.14; N 20.42.

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Benzimidazole 4 was obtained in 69% yield; mp 137-139°C (toluene). IR spectrum (KBr), ν, cm^{-1}: ν_{C-N} 1230, $\nu_{C=N}$ 1650, $\nu_{C\equiv C}$ 2200, ν_{NH} 3050. ^{1}H NMR spectrum (CD_3OD), δ, ppm: 2.23 (3H, s, CH_3); 5.70 (2H, s, CH_2); 6.98-7.26 (14H, m, H arom). Mass spectrum, *m/z* (I_{rel}, %): 365 (23), M^+ 364 (92), $[M-H]^+$ 363 (100), $[M-CH_3]^+$ 349 (9), $[(M-H) - CH_3CN]^+$ 322 (5), $[M-C_6H_5]^+$ 287 (27), $[M-CH_2C_6H_5]^+$ 273 (11), $[(M+H) - NCH_2C_6H_5]^+$ 260, 243 (13), $[(M+H) - C_9H_{10}N_2]^+$ 219 (6), 92 (13), $[CH_2C_6H_5]^+$ 91 (26), $[C_6H_5]^+$ 77 (5). Found, %: C 78.95; H 5.32; N 15.30. $C_{24}H_{20}N_4$. Calculated, %: C 79.09; H 5.53; N 15.37.

Benzimidazole 5 was obtained in 73% yield; mp 266-267°C (aqueous methanol). ^{1}H NMR spectrum (CD_3OD), δ, ppm: 2.15 (3H, s, CH_3); 2.25 (3H, s, CH_3); 5.39 (2H, s, CH_2); 7.10-7.45 (14H, m, H arom). Mass spectrum, *m/z* (I_{rel}, %): M^+ 364 (8), $[M-CH_2C_6H_5]^+$ 273 (11), $[M-NC(CH_3)C\equiv CC_6H_5]^+$ 222 (5), 93 (5), 92 (62), $[CH_2C_6H_5]^+$ 91 (100), 56 (9), 34 (13). Found, %: C 78.99; H 5.55; N 15.26. $C_{24}H_{20}N_4$. Calculated, %: C 79.09; H 5.53; N 15.37.

REFERENCES

1. N. A. Klyuev, M. V. Povstyanoi, and V. P. Gumennyi, *Khim. Geterotsikl. Soedin.*, 88 (1983).
2. M. V. Povstyanoi, V. P. Kruglenko, E. N. Fedosenko, and N. A. Klyuev, *Khim. Geterotsikl. Soedin.*, 234 (1990).
3. M. V. Povstyanoi, E. N. Fedosenko, and V. P. Kruglenko, *Ukr. Khim. Zh.*, No. 10, 1089 (1990).
4. M. V. Povstyanoi, V. P. Kruglenko, and V. M. Povstyanoi, *Khim. Geterotsikl. Soedin.*, 127 (2001).