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SELF-FIXATION OF RECYCLIZATION OF 2-PHENACYL-1H-BENZIMIDAZOLES HYDRAZONES

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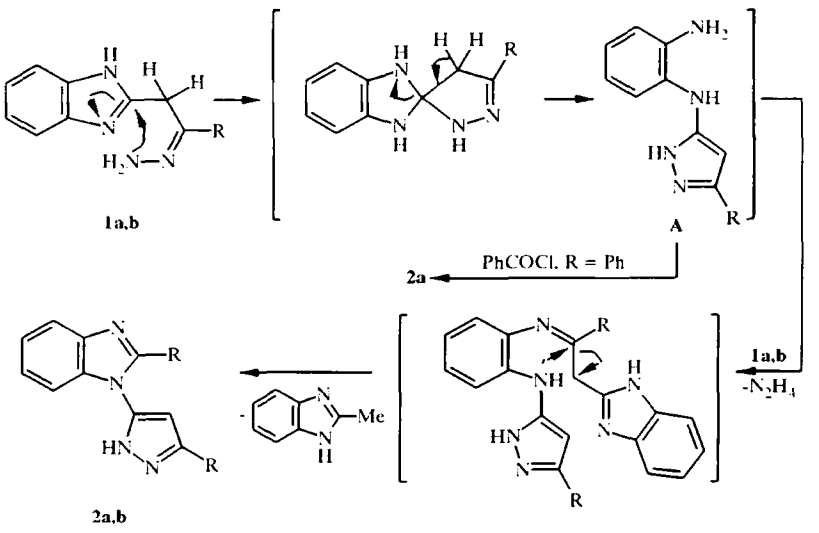
The formation of pyrazole **A** in the recyclization of 2-phenacyl-1H-benzimidazole hydrazone (**1a**) may be fixed by condensation with acylating reagents. In particular, a derivative of 1-[5(3)-pyrazolyl]-benzimidazole **2a** was obtained using benzoyl chloride [1].

We carried out the recyclization without an acylating reagent. The reaction proceeds under acid catalysis conditions and the major product in this case was also compound **2a**. This result may be seen as the condensation of expected product **A** with the starting hydrazone, possibly proceeding through intermediate formation of **B**.

Analogously, compound **1b** is converted into **2b**.

Thus, the products of the recyclization of 2-phenacyl-1H-benzimidazoles hydrazones are so reactive that they react upon formation under the reaction conditions with the starting compounds to give 1-[5(3)-pyrazolyl]-benzimidazoles.



a R = Ph, b R = 4-MeOC$_6$H$_4$

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2-Phenyl-1-[3(5)-phenylpyrazol-5(3)-yl]-1H-benzimidazole (2a). A mixture of **1a** (1 mol), *p*-toluenesulfonic acid (1.5 mmol), and dioxane (0.1 ml) was maintained for 4 h at 135°C. The mixture was then dissolved in 2-propanol (2 ml) at reflux and concentrated ammonium hydroxide (0.3 ml) was added. The product was separated by adding water to boiling solution until the onset of crystallization. The subsequent work-up and physicochemical indices of the products were in accord with those given in our previous work [1]. Yield of **2a** 49%.

2-(4-Methoxyphenyl)-1-[3(5)-(4-methoxyphenyl)pyrazolyl-5(3)-yl]-1H-benzimidazole (2b). A mixture of **1b** (1 mmol), hydrazine hydrochloride (1.5 mmol), and ethyleneglycol (1 ml) was maintained for 5 h at 135°C. Then, 2-propanol (1 ml), concentrated ammonium hydroxide (0.3 ml), and water (10 ml) were added. The resultant oil was converted into a powder by heating at reflux and stirring. Crystallization from 1:2 aqueous ethanol gave **2b** in 49% yield; mp 177-178°C. ^{1}H NMR spectrum (300 MHz, DMSO-d_6, TMS): 3.77 (3H, s, OCH_3); 3.81 (3H, s, OCH_3); 6.80 (1H, s, 4'-H); 6.97-7.78 (8H, m, $2C_6H_4$); 7.24-7.37 (3H, m, 4-H, 5-H, 6-H); 7.76 (1H, m, 7-H); 13.60 ppm (1H, m, NH). Found, %: C 72.60; H 5.13; N 14.28. $C_{24}H_{20}N_4O_2$. Calculated, %: C 72.71; H 5.09; N 14.13.

REFERENCES

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