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SYNTHESIS OF 5-(1H-BENZIMIDAZOL-2-YL)-1H-PYRAZOLO-[3,4-*b*]PYRIDINES BY THE *p*-(DIMETHYLAMINO)BENZALDEHYDE MODIFICATION OF HANTZSCH REACTION

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The reactions of p-(dimethylamino)benzaldehyde with 5-amino-3-methyl-1-phenylpyrazole and 2-RCOCH$_2$-1H-benzimidazoles have produced 5-(1H-benzimidazol-2-yl)-1H-pyrazolo[3,4-b]pyridines. The transformation includes the formation of compounds in accordance with a Hantzsch reaction containing a 1,4-dihydropyridine ring, and is completed by the aromatization either by the splitting off of N,N-dimethylanyline or oxidation. The splitting is produced by acetic acid and the oxidation by a nitrocompound.

Keywords: aldehydes, benzimidazoles, pyrazolo[3,4-*b*]pyridines, pyrazoles, Hantzsch reaction, aromatization, selectivity.

The synthesis of substituted pyridines in the Hantzsch reaction (cyclocondensation of an aldehyde with acetoacetic ester and ammonia followed by the oxidation of the resulting 1,4-dihydropyridines) is widely used in various modifications in organic synthesis [1-4]. If one boils *p*-(dimethylamino)benzaldehyde (**1**) in acetic acid with dimedone and ammonium acetate, the product containing a pyridine ring is as follows without the use of an oxidizing agent: the reaction occurs *via* a derivative of acridine with 10-[*p*-(dimethylamino)phenyl]-9,10-dihydropyridine moiety and is completed by the aromatization of the latter as a result of the splitting off of N,N-dimethylaniline [5]. If there is a 4-methoxy or 4-hydroxyphenyl substituent on atom C-10 in the 1,4-dihydropyridine moiety of these compounds, then the analogous splitting off of benzene, anisole, or phenol respectively occurs only on heating with mineral acids [6]. It is best to use the easy splitting off of N,N-dimethylaniline to obtain new compounds containing unsubstituted pyridine rings. By comparison with the usual synthesis methods, that approach has certain advantages: the process occurs in one stage and is simple to operate, being accessible and stable on storage and ease in dispensing the initial aldehyde **1**, together with high solubility of the resulting N,N-dimethylaniline, which eases the isolation of the target products from the reaction mixture.

Here I examine the scope for applying the new approach to examples of three-component reactions of the aldehyde **1**, 5-amino-3-methyl-1-phenylpyrazole (**2**), and 2-R-COCH$_2$-substituted 1H-benzimidazoles **3a-g**. Other researchers have reported [7-9] the use of type **2** aminopyrazoles in the synthesis of pyrazolopyridines; carbonyl components of type **3** have not been used in this reaction before, although it allows one to synthesize previously unknown benzimidazolyl-substituted pyrazolopyridines.

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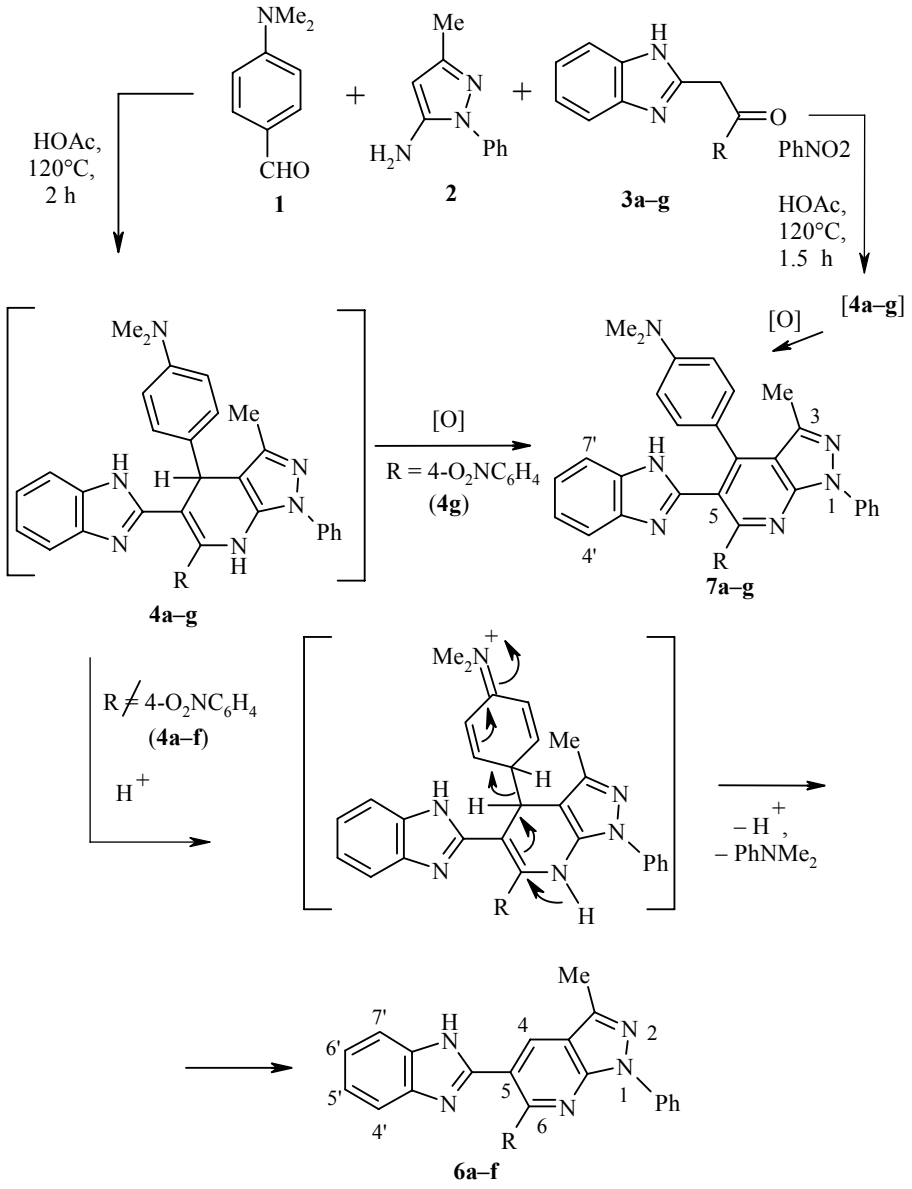

3–**7** **a** R = Me, **b** R = Ph, **c** R = 4-$MeOC_6H_4$, **d** R = 3,4,5-$(MeO)_3C_6H_2$,
e R = 4-BrC_6H_4, **f** R = 2-furyl, **g** R = 4-$O_2NC_6H_4$

The interaction of reagents **1**, **2**, and **3a-f** on boiling in acetic acid (method A) does not end with the formation of compounds **4a-f** containing the 1,4-dihydropyridine ring, but is instead accompanied by aromatization, which probably occurs by the C-protonized forms **5a-f**, which readily split off N,N-dimethylaniline and converted to the final products: 4-unsubstituted 5-(1H-benzimidazol-2-yl)-1H-pyrazolo[3,4-*b*]pyridines **6a-f** with yields of 69-95% (Table 1). The process occurs smoothly even with compound **3f**, which contains a furyl substituent and may be unstable in an acid medium. The aromatization due to oxidation (possibly by atmospheric oxygen) preserves the (dimethylamino)phenyl substituent and leads to the pyrazolo[3,4-*b*]pyridines **7a-f** to only a slight extent (as indicated by TLC). On the other hand, in the case of the benzimidazole **3g** (R = 4-$O_2NC_6H_4$), the conditions of method A lead to the oxidation predominating and the main product being pyrazolo[3,4-*b*]pyridine **7g** (yield 50%)

TABLE 1. Characteristics of Synthesized Compounds

Compound	Empirical formula	Found, % / Calculated, %			mp, °C	Yield, % (method)
		C	H	N		
6a	$C_{21}H_{17}N_5$	74.24 / 74.32	4.98 / 5.05	20.48 / 20.63	276.5-278	95 (A)
6b	$C_{26}H_{19}N_5$	77.53 / 77.79	4.95 / 4.77	17.28 / 17.44	313-315	80 (A)
6c	$C_{27}H_{21}N_5O$	75.02 / 75.16	4.78 / 4.91	16.07 / 16.23	276-278	89 (A)
6d	$C_{29}H_{25}N_5O_3$	70.68 / 70.86	5.32 / 5.13	14.09 / 14.25	262.5-264	85 (A
6e	$C_{26}H_{18}BrN_5$	64.87 / 65.01	3.95 / 3.78	14.43 / 14.58	330-331.5	95 (A)
6f	$C_{23}H_{15}N_5O$	73.48 / 73.64	4.21 / 4.38	17.56 / 17.89	251-252.5	69 (A)
7a	$C_{29}H_{26}N_6$	75.81 / 75.96	5.54 / 5.71	18.39 / 18.33	>350	52 (B); 46 (C)
7b	$C_{29}H_{28}N_6$	78.29 / 78.44	5.57 / 5.42	16.04 / 16.14	>350	75 (B)
7c	$C_{35}H_{30}N_6O$	76.23 / 76.34	5.36 / 5.49	15.18 / 15.26	>350	58 (B); 48 (C)
7d	$C_{37}H_{34}N_6O_3$	72.46 / 72.77	5.47 / 5.61	13.62 / 13.76	291-292.5	66 (B)
7e	$C_{34}H_{27}BrN_6$	68.08 / 68.12	4.63 / 4.54	13.94 / 14.02	>350	82 (B); 76 (C)
7f	$C_{32}H_{26}N_6O$	75.11 / 75.27	5.22 / 5.13	16.37 / 16.46	>350	56 (B); 29 (C)
7g	$C_{34}H_{27}N_7O_2$	72.06 / 72.20	4.66 / 4.81	17.21 / 17.33	328.5-330	50 (A); 71 (B); 69 (C)

The reactions of reagents **1**, **2**, and **3a-g** on the addition of nitrobenzene (oxidizing agent) is displaced towards the formation of products of type **7**. For example, with ratios for nitrobenzene: **1:2:3** of 5:1:1:1 (method B) that conversion line becomes preferred and the yields of products **7a-g** constitute 52-82%. Reduction of the amount of oxidizing agent by a factor 5 (method C) where compounds **3a,c,e-g** leads to reduction in the yield, particularly for compound **7f** (from 56 to 29%).

Increased electroacceptor capacity of the substituents R favors the formation of compounds **7a-g**; probably, the tendency of the intermediates **4** to oxidation increases with the acidity of the hydrogen atom in the γ-position of the 1,4-dihydropyridine fragment, but the better yield of the nitrophenyl-substituted product **7g** (71%), although higher than when nitrobenzene is used (50%) is less than the yields of the bromophenyl and even phenyl derivatives **7e** and **7b** (82 and 75%). Consequently, the initial compound **3g** is appreciably consumed in the oxidation even in the presence of nitrobenzene possibly because of the high oxidizing capacity of the nitrogroup.

The compositions and structures of the synthesized pyrazolo[3,4-*b*]pyridines of types **6** and **7** are confirmed by elemental analysis (Table 1) as well as ^{1}H NMR spectra (Table 2). Compounds **6a-f** are of the same structure type, as is implied by the chemical shifts of the signals for identically placed protons in the bicyclic fragments having only very small differences, which are quite regularly related to the nature of the substituents R. The same applies to compounds **7a-g**.

The ^{1}H NMR spectra of compounds of type **6** contain a distinct singlet signal for the H-4 proton on the weak-field side (8.63-8.76 ppm), which is absent from the spectra of compounds of type **7**. The signals from the *o*-, *m*-, and *p*-protons in the N-phenyl substituent appear separately in order of increasing field, with the *o*-protons at a fairly weak field (8.34-8.42 ppm), which is related to the descreening effect of the nitrogen atoms in the pyridine and pyrazole rings.

TABLE 2. ^{1}H NMR Spectra of Synthesized Compounds

Com-pound	δ, ppm (*J*, Hz)
6a	2.66 (3H, s, 3-CH_3); 2.99 (3H, s, 6-CH_3); 7.26-7.30 (2H, m, H-5',6'); 7.33 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.58 (3H, t, *J* = 7.5, $2H_{Ph}$-*m*, H-7'); 7.73 (1H, d, *J* = 7.8, H-4'); 8.34 (2H, d, *J* = 8.4, $2H_{Ph}$-*o*); 8.71 (1H, s, H-4); 12.86 (1H, s, NH*)
6b	2.68 (3H, s, 3-CH_3); 7.18-7.21 (2H, m, H-5',6'); 7.32-7.36 (4H, m, H_{NPh}-*p*, $3H_{CPh}$-*m*,*p*); 7.39-7.42 (1H, m, H-7'); 7.49 (2H, d, *J* = 7.5, $2H_{CPh}$-*o*); 7.59 (2H, t, *J* = 7.5, $2H_{NPh}$-*m*); 7.63-7.66 (1H, m, H-4'); 8.36 (2H, d, *J* = 7.2, $2H_{NPh}$-*o*); 8.73 (1H, s, H-4); 12.39 (1H, s, NH)
6c	2.64 (3H, s, 3-CH_3); 3.73 (3H, s, CH_3O); 6.87 (2H, d, *J* = 8.7, $2H_{Ar}$-*m*); 7.19-7.22 (2H, m, H-5',6'); 7.33 (1H, t, *J* = 7.5, H_{Ph}-*o*); 7.42 (2H, d, *J* = 8.1, $2H_{Ar}$-*m*, H-7'); 7.58 (2H, t, *J* = 8.1, $2H_{Ph}$-*m*); 7.64-7.67 (1H, m, H-4'); 8.37 (2H, d, *J* = 8.1, $2H_{Ph}$-*o*); 8.66 (1H, s, H-4); 12.42 (1H, s, NH)
6d	2.68 (3H, s, 3-CH_3); 3.44 (6H, s, $2CH_3O$-*m*); 3.65 (3H, s, CH_3O-*p*); 6.81 (2H, s, $2H_{Ar}$-*o*); 7.21-7.24 (2H, m, H-5',6'); 7.34 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.44-7.47 (1H, m, H-7'); 7.60 (2H, t, *J* = 8.1, $2H_{Ph}$-*m*); 7.67-7.70 (1H, m, H-4'); 8.42 (2H, d, *J* = 7.5, $2H_{Ph}$-*o*); 8.72 (1H, s, H-4); 12.47 (1H, s, NH)
6e	2.67 (3H, s, CH_3); 7.19-7.22 (2H, m, H-5',6'); 7.34 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.38 (2H, d, *J* = 9.0, $2H_{Ar}$-*m*); 7.44-7.46 (1H, m, H-7'); 7.53 (2H, d, *J* = 9.0, $2H_{Ar}$-*o*); 7.58 (2H, t, *J* = 7.5, $2H_{Ph}$-*m*); 7.62-7.66 (1H, m, H-4'); 8.34 (2H, d, *J* = 7.8, $2H_{Ph}$-*o*); 8.76 (1H, s, H-4); 12.50 (1H, s, NH)
6f	2.64 (3H, s, CH_3); 6.45 (1H, d, *J* = 3.3, H_{Het}-3); 6.54-6.55 (1H, m, H_{Het}-4); 7.25-7.27 (2H, m, H-5',6'); 7.35 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.53 (1H, d, *J* = 6.0, H-7'); 7.61 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 7.71 (1H, d, *J* = 8.7, H-4'); 7.75 (1H, d, *J* = 0.9, H_{Het}-5); 8.39 (2H, d, *J* = 8.4, $2H_{Ph}$-*o*); 8.63 (1H, s, H-4); 12.75 (1H, s, NH)
7a	2.06 (3H, s, 3-CH_3); 2.37 (3H, s, 6-CH_3); 2.84 [6H, s, $N(CH_3)_2$]; 6.58 and 7.14 (2×2H, two d, *J* = 9.0, $2H_{Ar}$-*o*, $2H_{Ar}$-*m*); 7.10-7.13 (2H, m, H-5',6'); 7.34 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.39-7.42 (1H, m, H-4'); 7.55-7.61 (3H, m, $2H_{Ph}$-*m*, H-4'); 8.31 (2H, d, *J* = 7.8, $2H_{Ph}$-*o*); 12.45 (1H, s, NH*)
7b	2.08 (3H, s, 3-CH_3); 2.85 [6H, s, $N(CH_3)_2$]; 6.60 (2H, d, *J* = 8.7, $2H_{Ar}$-*m*); 7.05-7.11 (2H, m, H-5',6'); 7.17 (2H, d, *J* = 8.4, $2H_{Ar}$-*o*); 7.19-7.26 (3H, m, $2H_{CPh}$-*m*, H_{CPh}-*p*); 7.26-7.29 (1H, m, H-7'); 7.34 (1H, t, *J* = 7.5, H_{NPh}-*p*); 7.44-7.49 (3H, m, $2H_{CPh}$-*o*, H-4'); 7.58 (2H, t, *J* = 7.8, $2H_{NPh}$-*m*); 8.34 (2H, d, *J* = 7.8, $2H_{NPh}$-*o*); 12.31 (1H, s, NH)
7c	2.05 (3H, s, 3-CH_3); 2.84 [6H, s, $N(CH_3)_2$]; 3.67 (3H, s, CH_3O), 6.58 (2H, d, *J* = 8.7, $2H_{4\text{-}Ar}$-*m*); 6.75 (2H, d, *J* = 8.7, $2H_{6\text{-}Ar}$-*m*); 7.07-7.09 (2H, m, H-5',6'); 7.13 (2H, d, *J* = 8.7, $2H_{4\text{-}Ar}$-*o*); 7.27-7.31 (1H, m, H-7'); 7.33 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.40 (2H, d, *J* = 8.7, $2H_{6\text{-}Ar}$-*o*); 7.48-7.50 (1H, m, H-4'); 7.58 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 8.34 (2H, d, *J* = 7.8, $2H_{Ph}$-2,6); 12.35 (1H, s, NH)
7d	2.04 (3H, s, 3-CH_3); 2.80 [6H, s, $N(CH_3)_2$]; 3.33 (6H, s, $2CH_3O$-*m*); 3.57 (3H, s, CH_3O-*p*); 6.56 (2H, d, *J* = 8.7, $H_{4\text{-}Ar}$-*m*); 6.73 (2H, s, C_6H_2); 7.05-7.09 (4H, m, H-5',6', $2H_{4\text{-}Ar}$-*o*); 7.22-7.25 (1H, m, H-7'); 7.33 (1H, t, *J* = 7.8, H_{Ph}-*p*); 7.40-7.43 (1H, m, H-4'); 7.59 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 8.38 (2H, d, *J* = 7.8, $2H_{Ph}$-*o*); 12.60 (1H, s, NH)
7e	2.08 (3H, s, 3-CH_3); 2.84 [6H, s, $N(CH_3)_2$]; 6.58 (2H, d, *J* = 8.7, $2H_{4\text{-}Ar}$-*m*); 7.08-7.10 (2H, m, H-5',6'); 7.15 (2H, d, *J* = 9.0, $2H_{4\text{-}Ar}$-*o*); 7.27-7.30 (1H, m, H-7'); 7.35-7.44 (5H, m, H_{Ph}-*p*, $4H_{6\text{-}Ar}$); 7.47-7.50 (1H, m, H-4'); 7.58 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 8.31 (2H, d, *J* = 7.8, $2H_{Ph}$-*o*); 12.38 (1H, s, NH)
7f	2.02 (3H, s, 3-CH_3); 2.84 [6H, s, $N(CH_3)_2$]; 5.75 (1H, d, *J* = 3.6, H_{Het}-3); 6.37-6.38 (1H, m, H_{Het}-4); 6.55 и 7.13 (2 × 2H, two d, *J* = 9.0, $4H_{4\text{-}Ar}$); 7.14-7.18 (2H, m, H-5',6'); 7.36 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.39-7.41 (1H, m, H-7'); 7.57-7.58 (1H, m, H-4'); 7.61 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 7.67 1H, d, *J* = 0.9, H_{Het}-5); 8.39 (2H, d, *J* = 7.8, $2H_{Ph}$-*m*); 12.69 (1H, s, NH)
7g	2.11 (3H, s, 3-CH_3); 2.84 [6H, s, $N(CH_3)_2$]; 6.60 (2H, d, *J* = 8.7, $2H_{4\text{-}Ar}$-*m*); 7.07-7.10 (2H, m, H-5',6'); 7.16 (2H, d, *J* = 9.0, $2H_{4\text{-}Ar}$-*o*); 7.28-7.30 (1H, m, H-7'); 7.36 (1H, t, *J* = 7.5, H_{Ph}-*p*); 7.47-7.50 (1H, m, H-4'); 7.58 (2H, t, *J* = 7.8, $2H_{Ph}$-*m*); 7.66 and 8.05 (2×2H, two d, *J* = 8.7, $4H_{6\text{-}Ar}$); 8.30 (2H, d, *J* = 8.1, $2H_{Ph}$-*o*); 12.46 (1H, s, NH)

* Undergoes deuterium exchange.

The signals from the 3-Me group in compounds of type **6** lie in the region of 2.64-2.68 ppm, while for compounds **7**, they lie at stronger fields (2.02-2.11 ppm), which is evidently due to the screening action of the adjacent *p*-(dimethylamino)phenyl substituent, which is deflected from the plane of the pyrazolopyridine ring because of steric factors related to the 5-benzimidazolyl substituent and the 3-Me group.

The 6-Me group in compound **6a** resonates at 2.99, while that of compound **7a** does so at 2.37 m.p., i.e., in a stronger field. The descreening action of the benzimidazole fragment in position 5 as regards this group is reduced by the steric influence from position 4.

In compounds **6** and even more in compounds **7**, steric interactions between the aromatic substituents in the pyridine ring of the pyrazolopyridine system prevent their having a coplanar position with respect to the plane of the latter. Consequently, the hydrogen atoms in substituent R located near the junction with the α-position of the position ring resonate at a stronger field than might be expected. Evidently, they to a considerable extent experience not descreening by the pyridine nitrogen atom but screening and steric displacement of the benzimidazole fragment. This is particularly evident in the case of the 2-furyl substituent: the signal from H_{Het}-3 is in an extremely strong field at 6.45 (compound **6f** or 5.75 ppm (compound **7f**), whereas the H_{Het}-4 signal is displaced to a weaker field: 6.54 (**6f**) or 6.37 ppm (**7f**).

The signals from protons H-4',7' in the benzimidazole ring appear separately, which indicates hindered migration of the protons between the nitrogen atoms. That is typical of benzimidazoles containing bulky heterocyclic substituents in position 2 [10].

Important information was obtained from the Overhauser effect of compounds **7g** and **6b**. The suppression of the signal from the 3-Me group in the first is not reflected in any of the other signals, including those from the 4-(dimethylamino)phenyl substituent (probably on account of its deviation from the plane of the pyrazolopyridine fragment). On the other hand, suppression of the 3-Me group signal of compound **6b** increases the integral intensity of the H-4 signal, while suppression of the latter signal is reflected in the first. These results confirm the spatially close position of the interacting nuclei, i.e., they indicate that reagents **1-3** react on the above scheme. This rules out the second possible mode of cyclocondensation, in which the aldehyde reacts on the aminogroup of the pyrazole **2** and the active methylene group of compound **3**, while the ring closure occurs as a result of the condensation of the C=O group in position **4** of the pyrazole ring. That transformation path would give rise to isomeric products differing in the arrangement of the substituents in the pyrazolo[3,4-*b*]pyridine fragment, and in particular having an α-unsubstituted position in the pyridine ring, which is spatially remote from the methyl group in the pyrazole ring.

These are examples of effective synthesis of previously unknown 4-unsubstituted 5-(1H-benzimidazol-2-yl)-1H-pyrazolo[3,4-*b*]pyridines of type **6** and show that the *p*-(dimethylamino)benzaldehyde modification of the reaction is promising.

EXPERIMENTAL

The reaction was monitored and the purity of the synthesized compounds was checked by TLC on Silufol UV-254 plates in a benzene-ethanol solvent system, 9:1, development in UV light. The ^{1}H NMR spectra were recorded with a Varian VXR-300 spectrometer (300 MHz) and a Varian Unity Plus-400 spectrometer (400 MHz, NOE experiment) in DMSO-d_6, standard TMS. All the compounds were dried for 5 h at 145°C before elemental analysis and the spectral studies.

5-(1H-benzimidazol-2-yl)-3,6-dimethyl-1-phenyl-1H-pyrazolo[3,4-*b*]pyridine (6a). A. A mixture of aldehyde **1** (0.149 g, 1 mmol), of aminopyrazole **2** (0.173 g, 1 mmol), of 2-acetonylbenzimidazole (**3a**) (0.174 g, 1 mmol), and 2 ml of glacial acetic acid was kept at 120°C for 2 h. Then 1 ml of water was added and the

mixture was heated with mixing to the start of crystallization. The solidified part was filtered off and the sediment of product **6a** was washed with a mixture of 2-propanol and water, 1:1. Yield 0.324 g (95%). The product was recrystallized from a pyridine–water mixture, 2:1.

Compounds **6b-f**, **7g** were made similarly from compounds **1**, **2**, and **3b-g** respectively. The **6f** product was recrystallized from a 1:1 pyridine–water mixture, while **7g** was recrystallized from a 5:1 dimethylformamide–water mixture.

5-(1H-benzimidazol-2-yl)-4-[4-(dimethylamino)phenyl]-3,6-dimethyl-1-phenyl-1H-pyrazolo[3,4-*b*]-pyridine (7a). B. A mixture of aldehyde **1** (0.149 g, 1 mmol), aminopyrazole **2** (0.173 g, 1 mmol), 2-acetonylbenzimidazole (**3a**) (0.174 g, 1 mmol), nitrobenzene (0.5 ml, 5 mmol), and 1 ml of glacial acetic acid was kept at 120°C for 1.5 h, and then we added 1 ml of water, 1 ml of 2-propanol, and boiled the mixture with stirring to the start of crystallization. The solidified mixture was filtered, and the deposit of product **7a** was washed with 2-propanol. Yield 0.238 g (52%). The product was recrystallized from dimethylformamide.

Compounds **7b-g** were made similarly respectively from reagents **1, 2,** and **3b-g** with nitrobenzene. To isolate the **7d** product, the reaction mixture was diluted with water (5 ml), evaporated to dryness, the residue was then boiled with 4 ml of acetone, and after cooling compound **7d** was filtered off, which was recrystallized from a 2:1 pyridine–water mixture. Compound **7e** was purified by crystallization from a 3:1 dimethylformamide–acetic acid mixture.

C. Compounds **7a, c,** and **e-g** were synthesized by the above method but with a nitrobenzene:**1**:**2**:**3** ratio of 1:1:1:1.

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