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SYNTHESIS AND AROMATIZATION OF 2-(3,6-DIARYL-2,5-DIHYDROPYRID-AZIN-4-YL)-1H-BENZIMIDAZOLES

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Treatment of 1,4-diaryl-2-(1H-benzimidazol-2-yl)butane-1,4-diones with hydrazine gives the previously unknown 2-(3,6-diaryl-2,5-dihydropyridazin-4-yl)-1H-benzimidazoles which are aromatized by oxidation with nitrous acid to give 2-[3,6-diarylpyridazin-4-yl]-1H-benzimidazoles.

Keywords: benzimidazole, hydrazine, 1,4-diketones, pyridazine.

We have previously reported the methodology of synthesizing novel benzimidazole derivatives based on 2-phenacyl-1H-benzimidazoles [1, 2]. In particular, C-alkylation of the latter using phenacyl bromides gave the 1,4-diaryl-2-(1H-benzimidazol-2-yl)butane-1,4-diones **1a-g** [3]. It is known [4] that 1,4-diketones can undergo cyclocondensation with hydrazine to form dihydropyridazines which are readily oxidized to pyridazines. Similar reactions of the 2-hetaryl-1,4-diketones of type **1** were unknown and are studied in this work.

We have found that the cyclocondensation of compounds **1a-g** with hydrazine hydrate occurs upon heating in a mixture of ethanol and pyridine and is accompanied by 1,3-migration of a proton from the methine group to the nitrogen atom in a direction leading not to compounds **2a-g** with a 1,3-dihydrobenzimidazole fragment but to the 2-(3,6-diaryl-2,5-dihydropyridazin-4-yl)-1-benzimidazoles **3a-g**. These products are obtained in 87-97% yields and are selectively oxidized by nitrous acid in pyridine (75-80ºC) undergoing aromatization to give 82-96% yields of the 2-(3,6-diarylpyridazin-4-yl)-1H-benzimidazoles **4a-g** (compounds **4a,d,g** crystallize in the form of molecular compounds with acetic acid with the compositions **4a**·AcOH, **4d**·AcOH, and **4g**·AcOH).

Evidently the formation of compounds of type **3** and their relative stability (they are not oxidized significantly when recording the NMR spectra in DMSO-d_6) is connected to the presence of an energetically favored conjugative chain in which the NH group of the dihydropyridazine ring is a donor and the benzimidazole fragment an acceptor of electrons. The conjugated chain constitutes a chromophoric system thus making compounds **3a-f** yellow and the compound **3g** with a nitrophenyl substituent orange colored. According to literature data, structural analogs of compound **3a** with a Ph or Me group in place of the benzimidazole fragment are colorless [5, 6] and with a C≡N group yellow [7]. We have found that, even with excess hydrochloric acid, the product **3a** forms the orange mono hydrochloride **5** in which the dihydropyridazine ring can be involved in the delocalization of the positive charge (see the limiting structure **5"**). Treatment of the salt **5** with ammonia regenerates the starting base **3a**.

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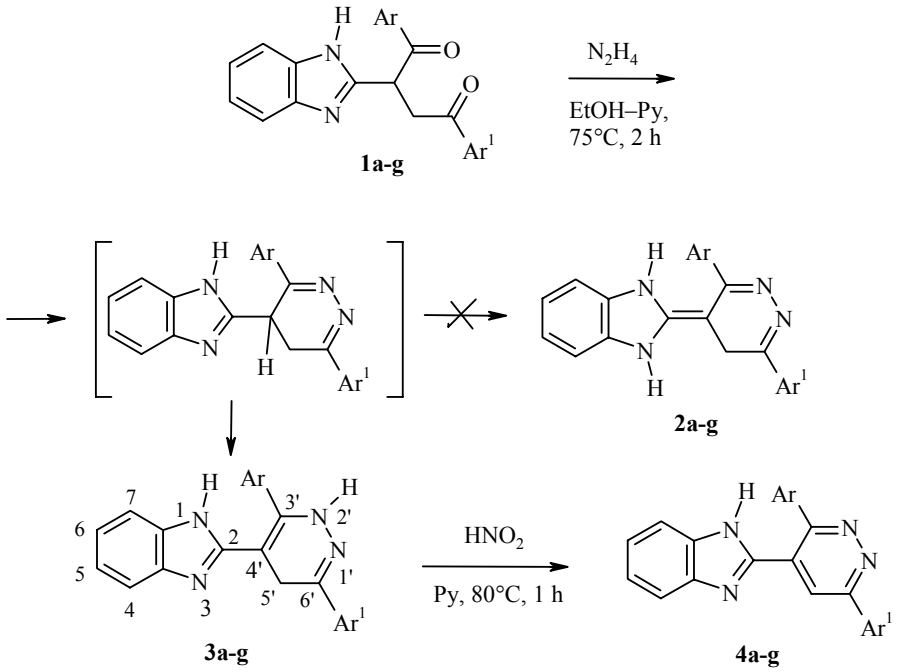
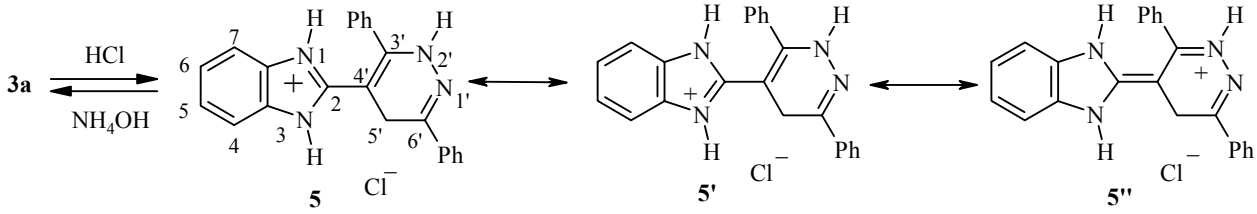


1–4 a Ar = Ar[1] = Ph, **b** Ar = 4-$MeOC_6H_4$, Ar[1] = Ph, **c** Ar = 3,4,5-$(MeO)_3C_6H_2$, Ar[1] = Ph, **d** Ar = 4-BrC_6H_4, Ar[1] = Ph, **e** Ar = Ph, Ar[1] = 3,4-$(MeO)_2C_6H_3$, **f** Ar = Ph, Ar[1] = 4-ClC_6H_4, **g** Ar = Ph, Ar[1] = 3-$O_2NC_6H_4$

The alternative structure for the product of cyclocondensation with hydrazine given by the formulae **2a-g** is less likely according to our data using the MOPAC quantum-chemical calculation program in the PM3 semiempirical approximation [8] which quite accurately yields the structure and molecular energy of organic compounds. The calculated values of the enthalpy of formation of structures **2a** and **3a** are



respectively 148.9 and 144.5 kcal/mol. The difference of 4.4 kcal/mol is quite significant and means that under equilibrium conditions in the gas phase at 25ºC the ratio of isomers **2a**:**3a** is about 1:1880. According to calculations for the compound **3a** molecule the most optimum conformation is found in which the coupling parts of both heterocyclic fragments are nonplanar because of steric hindrance and are placed at an angle of 45º to one another. In addition the azole H-1 atom is under the closest phenyl substituent which is turned by an angle of 82º to the plane of the aminovinyl fragment in the dihydropyridazine ring (see Figure 1).

The composition and structure of the synthesized compounds agreed with elemental analytical (Table 1) and ^{1}H NMR spectroscopic data (Table 2) and for compound **3a** also with chromatomass-analytical data.

The ^{1}H NMR spectra confirm that, in the molecules of compounds **3a-g**, both heterocyclic fragments represent a conjugated chain tending to a planar orientation but the steric hindrance brought about

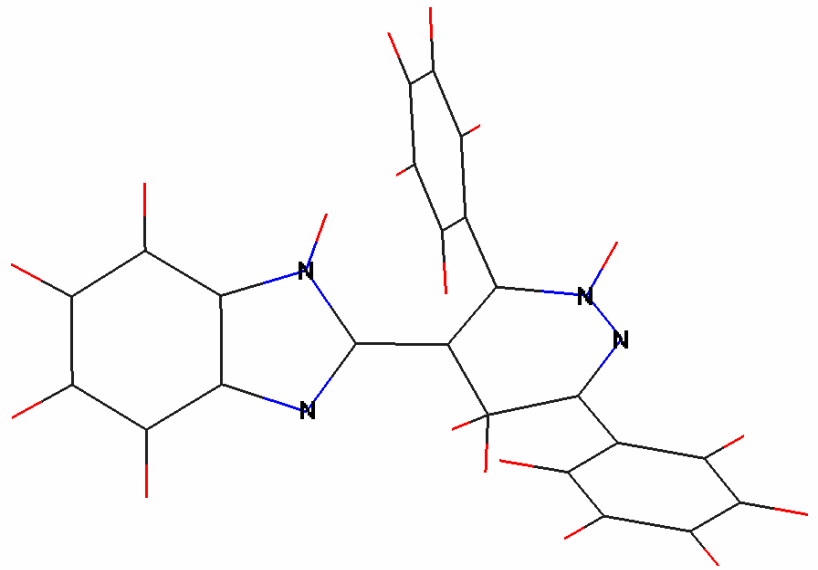


Figure 1. Basic conformation of the compound **3a** molecule calculated by the PM3 method.

TABLE 1. Characteristics of the Synthesized Compounds

Compound	Empirical formula	Found, % / Calculated, %			mp., °C	Yield, %
		C	H	N		
3a	$C_{23}H_{18}N_4$	78.67 / 78.83	5.24 / 5.18	15.78 / 15.99	235.0-237.0	90
3b	$C_{24}H_{20}N_4O$	75.59 / 75.77	5.35 / 5.30	14.65 / 14.73	272.0-273.5	88
3c	$C_{26}H_{24}N_4O_3$	70.78 / 70.89	5.55 / 5.49	12.56 / 12.72	216.5-218.0	97
3d	$C_{23}H_{17}BrN_4$	64.28 / 64.35	4.02 / 3.99	12.98 / 13.05	263.0-265.0	93
3e	$C_{25}H_{22}N_4O_2$	73.07 / 73.15	5.36 / 5.40	13.52 / 13.65	184.0-185.5	95
3f	$C_{23}H_{17}ClN_4$	71.56 / 71.78	4.38 / 4.45	14.47 / 14.56	232.5-234.0	88
3g	$C_{23}H_{17}N_5O_2$	69.72 / 69.86	4.27 / 4.33	17.63 / 17.71	232.0-233.5	87
4a ·AcOH	$C_{23}H_{16}N_4 \times C_2H_4O_2$	73.43 / 73.51	4.88 / 4.94	13.59 / 13.72	151.0-152.5	88
4b	$C_{24}H_{18}N_4O$	76.09 / 76.17	4.65 / 4.79	14.72 / 14.80	237.0-238.5	93
4c	$C_{26}H_{22}N_4O_3$	71.14 / 71.22	5.08 / 5.06	12.66 / 12.78	220.0-221.5	82
4d ·AcOH	$C_{23}H_{15}BrN_4 \times C_2H_4O_2$	64.59 / 64.65	3.47 / 3.54	12.97 / 13.11	235.0-236.5	96
4e	$C_{25}H_{20}N_4O_2$	73.43 / 73.51	4.91 / 4.94	13.63 / 13.72	242.5-244.0	85
4f	$C_{23}H_{15}ClN_4$	72.07 / 72.16	4.03 / 3.95	14.57 / 14.63	238.0-239.5	86
4g ·AcOH	$C_{23}H_{15}N_5O_2 \times C_2H_4O_2$	66.15 / 66.22	4.28 / 4.22	15.29 / 15.44	210.0-212.0	84
5	$C_{23}H_{18}N_4 \times HCl$*	71.14 / 71.40	5.27 / 4.95	14.23 / 14.48	190.0-191.5	89

* Found, %: Cl 9.16. Calculated, %: Cl 9.16.

is the reason for a near to perpendicular orientation of the Ar substituent relative to the plane of the aminovinyl part of the dihydropyridazine ring. Hence the signal for the *o*-protons of the Ar substituent is markedly shifted to high field and where Ar = Ph all five phenyl protons appear as a narrow multiplet at 7.39-7.40 ppm, i.e. have virtually the same values. On the other hand in the Ar^1 substituent at position 6 of the dihydropyridazine ring

TABLE 2. ^{1}H NMR Spectroscopic Parameters for the Synthesized Compounds

Com-pound	Chemical shifts, δ, ppm (*J*, Hz)
3a	3.67 (2H, s, CH_2); 7.02 (2H, m, H-5,6); 7.19 (1H, m, H-7); 7.39 (5H, m, 3'-C_6H_5); 7.40-7.46 (4H, m, H-4, $H_{Ph'}$-3,4,5); 7.82 (2H, d, *J* = 8.0, $H_{Ph'}$-2,6); 10.25 (1H, s, H-2', changed with D_2O slowly); 10.91 (1H, br. s, H-1*)
3b	3.68 (2H, s, CH_2); 3.79 (3H, s, OCH_3); 6.95 (2H, d, *J* = 8.0, H_{Ar}-3,5); 7.02 (2H, m, H-5,6); 7.25 (1H, m, H-7); 7.33 (2H, d, *J* = 8.0, H_{Ar}-2,6); 7.37-7.40 (2H, m, $H_{Ph'}$-4, H-4); 7.44 (2H, m, $H_{Ph'}$-3,5); 7.82 (2H, d, *J* = 8.0, $H_{Ph'}$-2,6); 10.20 (1H, s, H-2'); 10.80 (1H, br. s, H-1)
3c	3.63 (6H, s, 3"-,5"-OCH_3); 3.71 (5H, s, 4"-OCH_3, CH_2); 6.71 (2H, s, H_{Ar}-2,6); 7.04 (2H, m, H-5,6); 7.25 (1H, m, H-7); 7.39 (1H, m, $H_{Ph'}$-4); 7.45 (2H, m, $H_{Ph'}$-3,5); 7.50 (1H, m, H-4); 7.83 (2H, d, *J* = 8.0, $H_{Ph'}$-2,6); 10.24 (1H, s, H-2'); 10.88 (1H, br. s, H-1)
3d	3.70 (2H, s, CH_2); 7.04 (2H, m, H-5,6); 7.25 (1H, m, H-7); 7.34 (2H, d, *J* = 8.0, H_{Ar}-3,5); 7.39 (1H, m, $H_{Ph'}$-4); 7.44 (3H, m, $H_{Ph'}$-3,5, H-4); 7.58 (2H, d, *J* = 8.0, H_{Ar}-2,6); 7.82 (2H, d, *J* = 8.0, $H_{Ph'}$-2,6); 10.27 (1H, s, H-2'); 11.18 (1H, br. s, H-1)
3e	3.65 (2H, s, CH_2); 3.79 and 3.80 (3H and 3H, two s, 2OCH_3); 7.02 (3H, m, H-5,6, $H_{Ar'}$-5); 7.29-7.31 (2H, m, $H_{Ar'}$-6, H-7); 7.39-7.42 (6H, m, 3'-C_6H_5 + $H_{Ar'}$-2); 7.47 (1H, m, H-4); 10.15 (1H, s, H-2'); 10.91 (1H, br. s, H-1)
3f	3.69 (2H, s, CH_2); 7.02 (2H, m, H-5,6); 7.30 (1H, m, H-7); 7.39-7.45 (6H, m, 3'-C_6H_5, H-4); 7.50 and 7.83 (2 x 2H, two d, *J* = 8.0, 4H_{Ar}); 10.30 (1H, s, H-2'); 10.93 (1H, br. s, H-1)
3g	3.78 (2H, s, CH_2); 7.04 (2H, m, H-5,6); 7.21 (1H, m, H-7); 7.40-7.46 (6H, m, 3'-C_6H_5, H-4); 7.73 (1H, m, $H_{Ar'}$-5); 8.21 (2H, m, $H_{Ar'}$-4,6); 8.57 (1H, s, $H_{Ar'}$-2); 10.43 (1H, s, H-2'); 10.95 (1H, br. s, H-1)
4a·AcOH	1.90 (3H, s, $COCH_3$); 7.23 (2H, m, H-5,6); 7.38 (2H, m, H_{Ph}-3,5); 7.42 (1H, m, H_{Ph}-4); 7.51 (3H, m, H_{Ph}-2,6, H-7); 7.55-7.65 (4H, m, $H_{Ph'}$-3,4,5, H-4); 8.31 (2H, d, *J* = 8.0, $H_{Ph'}$-2,6); 8.57 (1H, s, H-5'); 11.96 (1H, br. s, OH); 12.77 (1H, br. s, H-1*)
4b	3.76 (3H, s, $COCH_3$); 6.95 and 7.48 (2H and 2H, two d, *J* = 8.0, H_{Ar}-2,3,5,6); 7.24 (2H, m, H-5,6); 7.52 (1H, m, H-7); 7.55–7.62 (3H, m, $H_{Ph'}$-3,4); 7.68 (1H, m, H-4); 8.29 (2H, d, *J* = 7.0, $H_{Ph'}$-2,6); 8.50 (1H, s, H-5'); 12.72 (1H, br. s, H-1)
4c	3.49 (6H, s, 3"-,5"-OCH_3); 3.67 (3H, s, 4"-OCH_3); 6.84 (2H, s, H_{Ar}-2,6); 7.26 (2H, m, H-5,6); 7.53 (1H, m, H-7); 7.60 (3H, m, $H_{Ph'}$-3,4,5); 7.70 (1H, m, H-4); 8.31 (2H, d, *J* = 7.0, $H_{Ph'}$-2,6); 8.57 (1H, s, H-5'); 12.73 (1H, br. s, H-1)
4d·AcOH	1.90 (3H, s, $COCH_3$); 7.23 (2H, m, H-5,6); 7.48 (2H, d, *J* = 8.0, H_{Ar}-3,5); 7.53-7.67 (7H, m, H-4,7, $H_{Ph'}$-3,4,5, H_{Ar}-2,6); 8.30 (2H, d, *J* = 7.0, $H_{Ph'}$-2,6); 8.59 (1H, s, H-5'); 11.95 (1H, br. s, OH); 12.86 (1H, br. s, H-1)
4e	3.85 and 3.90 (3H and 3H, two s, 2OCH_3); 7.16 (1H, d, *J* = 8.5, $H_{Ar'}$-5); 7.20-7.25 (2H, m, H-5,6); 7.36 (2H, m, H_{Ph}-3,5); 7.39 (1H, m, H_{Ph}-4); 7.48-7.51 (3H, m, H_{Ph}-2,6, H-7); 7.65 (1H, d, *J* = 7.0, H-4); 7.91 (1H, d, *J* = 8.5, $H_{Ar'}$-6); 7.92 (1H, s, $H_{Ar'}$-2); 8.55 (1H, s, H-5'); 12.76 (1H, br. s, H-1)
4f	7.25 (2H, m, H-5,6); 7.39 (3H, m, H_{Ph}-3,4,5); 7.50 (2H, d, *J* = 7.0, H_{Ph}-2,6); 7.56 (1H, m, H-7); 7.64-7.74 (3H, m, $H_{Ar'}$-3,5, H-4); 8.35 (2H, d, *J* = 8.5, $H_{Ar'}$-2,6); 8.60 (1H, s, H-5'); 12.76 (1H, br. s, H-1)
4g·AcOH	1.90 (3H, s, $COCH_3$); 7.24 (2H, m, H-5,6); 7.40 (2H, m, H_{Ph}-3,5); 7.44 (1H, m, H_{Ph}-4); 7.53 (2H, d, *J* = 7.0, H_{Ph}-2,6); 7.62 (2H, m, H-4,7); 7.92 (1H, m, $H_{Ar'}$-5); 8.42 (1H, d, *J* = 7.0, $H_{Ar'}$-6); 8.78 (1H, d, *J* = 7.0, $H_{Ar'}$-4); 8.79 (1H, s, H-5'); 9.12 (1H, s, $H_{Ar'}$-2); 11.93 (1H, br. s, OH); 12.78 (1H, br. s, H-1)
5	3.84 (2H, s, CH_2); 7.38 (2H, m, H-5,6); 7.46-7.59 (10H, m, 3'-C_6H_5, $H_{Ph'}$-3,4,5, H-4,7); 7.91 (2H, d, *J* = 7.0, $H_{Ph'}$-2,6); 11.24 (1H, s, H-2'); 13.63 (2H, br. s, H-1,3)

* Undergoes deuterium exchange.

steric hindrance does not occur and so it experiences the deshielding effect of the nearby ring nitrogen atom. As a result the *o*-protons signals are shifted to low field. Steric hindrances at the nitrogen atom of the benzimidazole fragment slow down processes of a mutual intermolecular proton transfer and this is reflected in the difference in position of the H-4 and H-7 signals in the spectrum. The first is found at 7.39-7.50 and the second at 7.19-7.30 ppm. We have previously observed features related to the orthogonal orientation of the aryl substituent relative to the heterocyclic plane and hindrance to proton exchange between benzimidazole nitrogen atoms in the spectra of structural analogs of compounds **3a-g** containing a partially hydrogenated pyrimidine or 1,4-benzothiazine ring [9, 10] in place of the pyridazine ring. When compared with the H-1 signal at 10.80-11.18 ppm the H-2' signal in **3a-g** is at higher field (10.15-10.43 ppm), it is less broadened, and its intensity is decreased less significantly after addition of D_2O. The protons of the CH_2 groups appear as a singlet signal at 3.65-3.78 ppm (in the starting compounds **1a-g** they are diastereotopic and appear as two double doublets [3]). The indicated signals for H-1,2' and the CH_2 groups are quite systematically shifted to low field with an increase in the electron-acceptor properties of the Ar and Ar^1 substituents.

In the ^{1}H NMR spectrum of salt **5** all of the signals are shifted to low field when compared to the signals for the base **3a**. Hence the signal for the azine amino group appears at 11.23 and the signals for H-1 and H-3 at 13.63 ppm as a broadened two-proton singlet.

The ^{1}H NMR spectra of compounds **4a-g** also agree well with the structures indicated in the scheme above. When compared with a similar substituent in compounds of type **3** the Ar substituent experiences less steric hindrance and undergoes a deshielding effect due to the nearby pyridazine nitrogen atom. In particular, where Ar = Ph (see the spectrum of **4a**·AcOH) the *o*-protons of the phenyl ring resonate at lower field than the *m*- and *p*-protons but with a lesser low-field shift than the *o*-protons of the substituent Ar^1 = Ph since they are influenced by the effect of the benzimidazole fragment. The H-5' signal appears in the range 8.50-8.79 ppm, consistently shifted to low field with increasing electron-acceptor properties of the Ar and Ar^1 substituents. In the spectra of the compounds **4a**·AcOH, **4d**·AcOH, and **4g**·AcOH the OH group proton signal of the acetic acid appears separately to the H-1 of the benzimidazole fragment and this excludes their salt forming nature (based on benzimidazole being more basic than pyridazine). The H-1 signal in the spectra of **4a**·AcOH, **4b,c**, and **4d**·AcOH (Ar^1 = Ph) is found at 12.72-12.86 ppm and shifted to low field with increasing electron-acceptor properties in the Ar substituent and in compounds **4a**·AcOH, **4e,f**, and **4g**·AcOH (Ar = Ph) at 12.76-12.78 ppm, i.e. virtually independent of the nature of the Ar^1 substituent due to the absence of a conjugated link.

Only unsubstituted 2-(pyridazin-4-yl)-1H-benzimidazole and its 3',6'-dimethyl-substituted homolog which were prepared by cyclocondensation of *o*-phenylenediamine with pyridazine-4-carboxylic acid derivatives were reported previously [11]. Information about 2-(dihydropyridazin-4-yl)-1H-benzimidazoles of type **3** is generally absent.

Hence in this work we propose a method for the synthesis of the previously unknown 2-(dihydropyridazin-4-yl)-1H-benzimidazoles which, in conjunction with subsequent product aromatization by nitrous acid, is a novel method for preparing the corresponding 2-(pyridazin-4-yl)-substituted products. An important feature of the proposed methods is the high yields of the target materials (82-97%).

EXPERIMENTAL

Monitoring of the course of the reaction and purity of the synthesized compounds was carried out by TLC on Silufol UV-254 plates in the solvent system benzene–ethanol, (9:1) and revealed in UV light. The ^{1}H NMR spectra of the compounds were recorded on a Bruker Avance DRX 500 (500 MHz) spectrometer in DMSO-d_6 using TMS as standard. Chromatomass-spectrometric analysis was performed by high resolution

liquid chromatography on an Agilent 1100 instrument fitted with an Agilent LC/MSD SL mass detector. Before determining the elemental analysis and spectroscopic investigation the compounds **3a-g, 4a**·AcOH, **4d**·AcOH, **4g**·AcOH, and **5** were dried for 20 h at 40-45ºC and the rest for 4 h at 125ºC.

2-(3,6-Diphenyl-2,5-dihydropyridazin-4-yl)-1H-benzimidazole (3a). A mixture of compound **1a** (0.354 g, 1 mmol), pyridine (0.5 ml), ethanol (2 ml), hydrazine hydrate (80%, 0.4 ml), and glacial acetic acid (2 drops) was held at 70-75ºC for 2 h under an argon atmosphere. The cooled product was filtered off and the precipitate was washed with ethanol and dried to give analytically pure **3a** (0.314 g). Chromato-mass spectrometric analysis: purity > 99%. Found, M+1 = 351. $C_{23}H_{18}N_4$. Calculated, M = 350.

Compounds 3b-g were prepared similarly from compounds **1b-g** respectively.

2-(3,6-Diphenylpyridazin-4-yl)-1H-benzimidazole (4a). A mixture of compound **3a** (0.175 g, 0.5 mmol), sodium nitrite (0.069 g, 1 mmol), pyridine (2 ml), glacial acetic acid (10 drops), and water (5 drops) was stirred at 80ºC for 15 min, water (5 drops) was added, the product was stirred, heated for 15 min, and water (2 ml) was again added. The reaction mixture was evaporated under reflux to a volume of 1 ml, diluted initially with glacial acetic acid (1 ml), and then with water (1 ml). The cooled product was filtered and the precipitate was washed with 2-propanol and dried to give analytically pure **4a**·AcOH.

Product 4g·AcOH was prepared similarly from compound **3g**.

Product 4c was obtained from compound **3c** but acetonitrile (1 ml) was used in the separation instead of acetic acid (1 ml).

Products 4b,d-f were synthesized similarly from compounds **3b,d-f** respectively. They crystallized just by dilution of the reaction mixture with water (2 ml) and compounds **4b,e,f** were obtained in an analytically pure state. The product **4d** was further purified by crystallization from acetic acid to form **4d**·AcOH.

2-(3',6''-Diphenyl-2',5'-dihydropyridazin-4-yl)-3H-benzimid-1-azolium Chloride (5). A mixture of compound **3a** (0.7 g, 2 mmol), acetone (1.5 ml), concentrated hydrochloric acid (0.5 ml, 5 mmol), and water (1.5 ml) was refluxed with stirring for 1-2 min. The cooled product was filtered off and the precipitate was washed with 2-propanol and dried to give analytically pure salt **5** (0.766 g).

Regeneration of base 3a from salt 5. A mixture of salt **5** (0.2 g), acetone (1.5 ml), and 20% aqueous ammonia solution (0.5 ml) was heated to reflux with stirring and then gradually diluted with water (4 ml). The cooled product was filtered to give compound **3a** (0.149 g, 82%) which was identical to the sample prepared in the scheme reported above for synthesizing product **3a** (TLC data and mixed melting point).

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