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CYCLOADDITION OF 2-ACYLMETHYL-1H-BENZIMIDAZOLES TO 4-ARYLIDENE-1,3-OXAZOL-5-ONES

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Cycloaddition of 2-phenacyl-1H-benzimidazole to 2-phenyl- or 2-methyl-4-arylidene-1,3-oxazol-5-ones occurs regioselectively to form the previously unknown N-(3-aryl-4-benzoyl-1-oxo-1,2,3,4-tetrahydropyrido[1,2-a]benzimidazol-2-yl)benz- and -acetamides. The analogous cycloaddition of the 2-acetonyl-1H-benzimidazole is complicated by prototropic isomerization and leads to the corresponding 1,2,3,5-tetrahydropyrido[1,2-a]benzimidazole.

Keywords: azlactones, benzimidazoles, pyrido[1,2-*a*]benzimidazoles, [3+3]cycloaddition, isomerization, regioselectivity.

Pyrido[1,2-*a*]benzimidazoles possess a broad spectrum of biological activity [1-6], have fluorescence properties [7], and are a component of light sensitive materials [8, 9]. We hoped to prepare previously unavailable derivatives on the basis of the reaction of 2-acylmethyl-1H-benzimidazoles of type **1** with 4-arylidene-1,3-oxazol-5-ones (azlactones) of type **2** and **3** for the following reasons. 2-Aminobenzimidazole reacts regioselectively with azlactones to give pyrimido[1,2-*a*]benzimidazoles, in such a way that the exocyclic amino group is acylated as a result of opening of the oxazole ring and the endocyclic by addition to the arylidene fragment [10]. It is advisable to use compounds of type **1** as the 1,3-dinucleophile component. In the first place 2-phenacyl-1H-bezimidazole **1a** is acylated by carboxylic acid anhydrides at the active methylene group [11]. In the second, azlactones are analogs of cyclic anhydrides which react with active methylene compounds to form C-acylation products [12].

We have found that the reaction of compound **1** with azlactones **2a-f, 3a-c** indeed occurs efficiently via the cycloaddition of a 1,3-dinucleophile to a 1,3-dielectrophile. However, the products of the expected structure **4** or any kind of prototropic isomers were not formed. A contrary scheme of regioselective construction or the ring is achieved *via* acylation at a benzimidazole nitrogen atom (possible by the intermediate formation of compound **5**) and Michael addition of the methylene group to the arylidene fragment to give the previously unknown N-(3-aryl-4-benzoyl-1-oxo-1,2,3,4-tetrahydropyridino[1,2-*a*]benzimidazol-2-yl)benz- and -acetamides **6a-f, 7a-c**.

The reaction was carried out by refluxing in pyridine and is complete after 1 h. The product yields were 70-95%.

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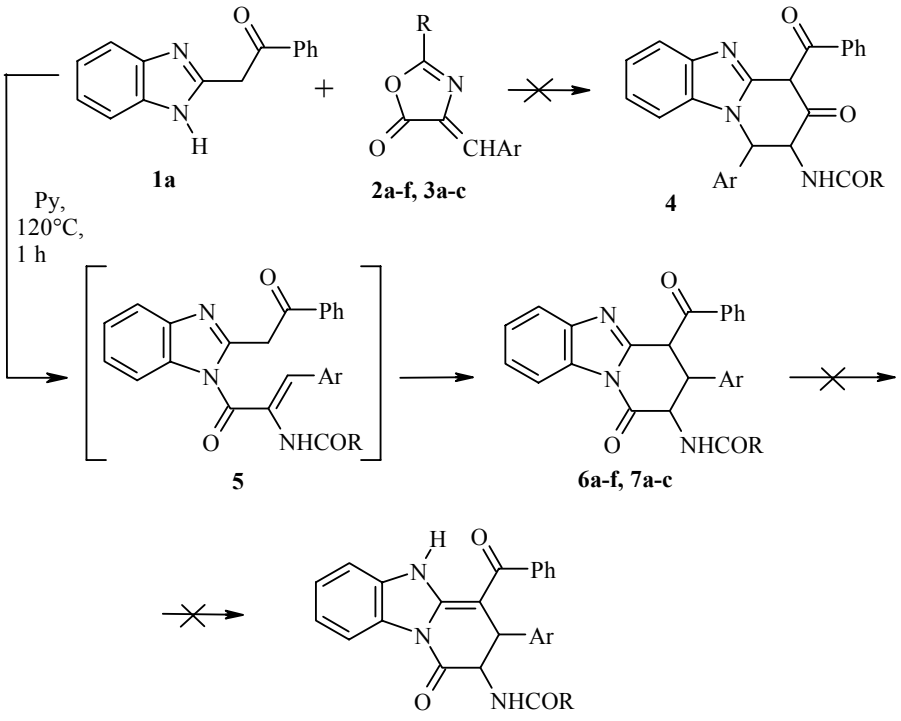
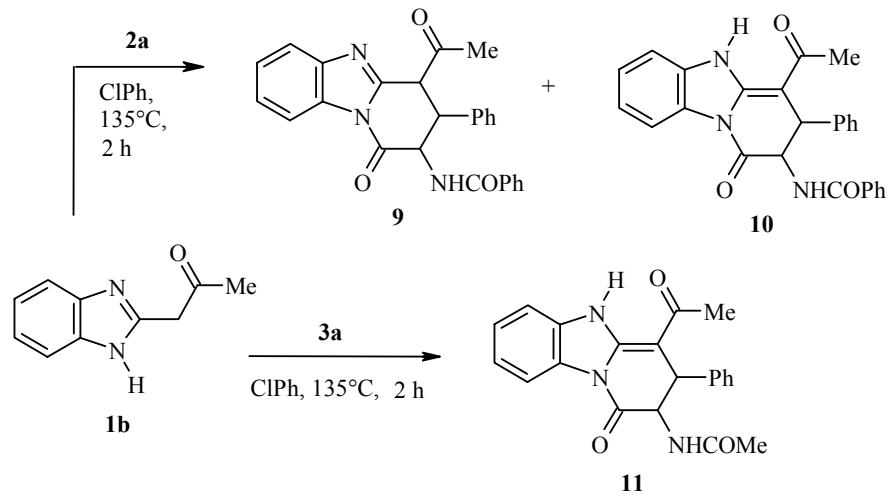
It should be noted that the starting compound **1a** exists in solutions in equilibrium with an enamino ketone tautomeric form [13] but the obtained products **6** and **7** do not show a similar tendency to isomerize to form **8**.



1a **2a-f, 3a-c** **4** **5** **6a-f, 7a-c** **8**

2, **6** R = Ph; **3**, **7** R = Me; **2**, **3**, **6**, **7** **a** Ar = Ph, **b** Ar = 4-$MeOC_6H_4$, **c** Ar = 4-ClC_6H_4,
2, 6 d Ar = 4-$O_2NC_6H_4$, **e** Ar = 3-Py, **f** Ar = 4-Py

On the other hand, the reaction of 2-acetonyl-1H-benzimidazole **1b** with the azlactones **2a** and **3a** occurs by the route revealed for compound **1a** but is complicated by a prototropic isomerization. The reaction can be carried out in pyridine or in chlorobenzene but the latter solvent is more convenient to use in the separation of the products from the reaction mixtures. Cycloaddition of the azlactone **2a** gave the expected compound **9** and its enamino ketone tautomer **10**. The cycloaddition of azlactone **3a** gave only the isomerization product **11**.



9 **10** **1b** **11**

Compounds **9-11** are fully stable in DMSO-d_6 solution for 3 h at 20-25ºC. Evidently the shift of the proton between the carbon and nitrogen atoms in positions 4 and 5 is hindered and is possible only at higher temperatures. Such a prototropic feature is likely due to the limit to conformational mobility of the structural fragment $C_{(4)}$–$C_{(4a)}$–$N_{(5)}$, the central atom of which is situated at the junction of two condensed rings. This also does not exclude the contribution of steric hindrance arising between the substituent in positions 2, 3, and 4.

TABLE 1. Characteristics of Compounds Synthesized

Compound	Empirical formula	Found, % / Calculated, %			mp, °C	Yield, %
		C	H	N		
6a	$C_{31}H_{23}N_3O_3$	76.52 / 76.69	4.48 / 4.77	8.48 / 8.65	293-294.5	95
6b	$C_{32}H_{25}N_3O_4$	74.46 / 74.55	4.78 / 4.89	8.03 / 8.15	316-317.5	85
6c	$C_{31}H_{22}ClN_3O_3$	71.58 / 71.61	4.32 / 4.26	8.09 / 8.08	302.5-304	89
6d	$C_{31}H_{22}N_4O_5$	70.02 / 70.18	4.29 / 4.18	10.39 / 10.56	306-307.5	85
6e	$C_{30}H_{22}N_4O_3$	73.89 / 74.06	4.43 / 4.56	11.35 / 11.52	263.5-265	70
6f	$C_{30}H_{22}N_4O_3$	74.03 / 74.06	4.49 / 4.56	11.34 / 11.52	296-294.5	84
7a	$C_{26}H_{21}N_3O_3$	73.62 / 73.74	5.16 / 5.00	9.79 / 9.92	290-291.5	90
7b	$C_{27}H_{23}N_3O_4$	71.48 / 71.51	5.28 / 5.11	9.17 / 9.27	260-261.5	85
7c	$C_{26}H_{20}ClN_3O_3$	68.15 / 68.20	4.28 / 4.40	9.09 / 9.18	270-271.5	88
9	$C_{26}H_{21}N_3O_3$	73.59 / 73.74	5.06 / 5.00	9.81 / 9.92	285-286.5	48
10	$C_{26}H_{21}N_3O_3$	73.57 / 73.74	5.05 / 5.00	9.77 / 9.92	217.5-219	26
11	$C_{21}H_{19}N_3O_3$	69.63 / 69.79	5.17 / 5.30	11.53 / 11.63	236-237	62

TABLE 2. IR and ^{1}H NMR Spectroscopic Characteristics of Compounds Synthesized

Compound	IR spectrum, cm^{-1} (C=S, C=O N-H)	^{1}H NMR spectrum, δ, ppm (*J*, Hz)
1	2	3
6a	1615, 1660, 1680, 1740, 3349, 3405	4.48 (1H, m, H-3); 5.70 (1H, br. m, H-2); 6.35 (1H, d, *J* = 11.7, H-4); 7.09 (1H, t, *J* = 7.2, 3-C_6H_5: H-4); 7.20 (2H, t, *J* = 7.2, 3-C_6H_5: H-3,5); 7.39-7.50 (6H, m, H-7,8 + 3-C_6H_5: H-2,6 + $NCOC_6H_5$: H-3,5); 7.50-7.57 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.65-7.67 (4H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6); 7.98 (2H, d, *J* = 7.5, 4-COC_6H_5: H-2,4); 8.21 (1H, d, *J* = 7.5, H-9); 8.94 (1H, br. m, NHCO, hindered exchange with D_2O)
6b	1610, 1645, 1680, 1735, 3340	3.60 (3H, s, OCH_3); 4.40 (1H, m, H-3); 5.73 (1H, br. m, H-2); 6.29 (1H, d, *J* = 12.0, H-4); 6.76 and 7.29 (2 + 2H, two d, *J* = 8.4, $C_6H_4OCH_3$); 7.40-7.48 (4H, m, H-7,8 + $NCOC_6H_5$: H-3,5); 7.51-7.59 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.65-7.69 (4H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6); 7.99 (2H, d, *J* = 7.2, 4-COC_6H_5: H-2,6); 8.20 (1H, d, *J* = 7.5, H-9); 8.91 (1H, br. m, NHCO)
6c	1620, 1650, 1680, 1740, 3340	4.47 (1H, m, H-3); 5.71 (1H, br. m, H-2); 6.35 (1H, d, *J* = 12.0, H-4); 7.29 (2H, d, *J* = 8.4, 4-ClC_6H_4: H-2,6); 7.39-7.48 (6H, m, H-7,8 + $NCOC_6H_5$: H-3,5 + 4-ClC_6H_4: H-4,5); 7.52-7.60 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.66-7.73 (4H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6); 7.99 (2H, d, *J* = 7.5, 4-COC_6H_5: H-2,6); 8.21 (1H, d, *J* = 7.2, H-9); 8.97 (1H, br. m, NHCO)

TABLE 2. (continued)

1	2	3
6d	1615, 1660, 1690, 1755, 3320	4.65 (1H, m, H-3); 5.80 (1H, br. m, H-2); 6.46 (1H, d, *J* = 12.6, H-4); 7.40-7.51 (4H, m, H-7,8 + $NCOC_6H_5$: H-3,5); 7.55-7.60 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.68 (6H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6 + 4-$NO_2C_6H_4$: H-2,6); 8.00 (2H, d, *J* = 7.2, 4-COC_6H_5: H-2,6); 8.12 (2H, d, *J* = 8.4, 4-$NO_2C_6H_4$: H-3,5); 8.21 (1H, d, *J* = 7.2, H-9); 9.02 (1H, br. m, NHCO)
6e	1645, 1680, 1725, 3360	4.50 (1H, m, H-3); 5.82 (1H, br. m, H-2); 6.42 (1H, d, *J* = 12.0, H-4); 7.24-7.28 (1H, m, C_5H_4N: H-5); 7.39-7.50 (4H, m, H-7,8 + $NCOC_6H_5$: H-3,5); 7.52-7.60 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.66-7.72 (4H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6); 7.87 (1H, d, *J* = 7.8, C_5H_4N: H-4); 8.01 (2H, d, *J* = 7.5, COC_6H_5: H-2,6); 8.22 (1H, d, *J* = 7.2, H-9); 8.32 (1H, d, *J* = 4.5, C_5H_4N: H-6); 8.53 (1H, s, C_5H_4N: H-2); 9.00 (1H, br. m, NHCO)
6f	1655, 1680, 1745, 3290, 3490	4.50 (1H, m, H-3); 5.77 (1H, br. m, H-2); 6.39 (1H, d, *J* = 11.7, H-4); 7.38-7.48 (6H, m, H-7,8 + $NCOC_6H_5$: H-3,5 + C_5H_4N: H-2,6); 7.52-7.60 (3H, m, 4-COC_6H_5: H-3,5 + $NCOC_6H_5$: H-4); 7.66-7.73 (4H, m, H-6 + 4-COC_6H_5: H-4 + $NCOC_6H_5$: H-2,6); 8.01 (2H, d, *J* = 7.5, COC_6H_5: H-2,6); 8.21 (1H, d, *J* = 7.2, H-9); 8.42 (2H, d, *J* = 5.4, C_5H_4N: H-3,5); 8.99 (1H, br. m, NHCO)
7a	1610, 1675, 1690, 1745, 3315	1.72 (3H, s, CH_3); 4.29 (1H, m, H-3); 5.33 (1H, br. m, H-2); 6.22 (1H, d, *J* = 12.3, H-4); 7.14 (1H, t, *J* = 7.2, 3-C_6H_5: H-4); 7.23 (2H, t, *J* = 7.2, 3-C_6H_5: H-3,5); 7.32 (2H, d, *J* = 7.2, 3-C_6H_5: H-2,6); 7.36-7.47 (2H, m, H-7,8); 7.53 (2H, t, *J* = 7.8, COC_6H_5: H-3,5); 7.63-7.68 (2H, m, H-6 + 4-COC_6H_5: H-4); 7.94 (2H, d, *J* = 7.5, 4-COC_6H_5: H-2,4); 8.19 (1H, d, *J* = 7.5, H-9); 8.35 (1H, br. d, *J* = 6.9, NHCO, hindered exchange with D_2O)
7b	1620, 1670, 1685, 1740, 3310	1.73 (3H, s, CH_3); 3.65 (3H, s, CH_3O); 4.22 (1H, m, H-3); 5.26 (1H, br. m, H-2); 6.18 (1H, d, *J* = 12.3, H-4); 6.79 and 7.23 (2 + 2H, two d, *J* = 8.4, $C_6H_4OCH_3$); 7.36-7.46 (2H, m, H-7,8); 7.54 (2H, t, *J* = 7.5, COC_6H_5: H-3,5); 7.62-7.69 (2H, m, H-6 + 4-COC_6H_5: H-4); 7.95 (2H, d, *J* = 7.8, 4-COC_6H_5: H-2,6); 8.18 (1H, d, *J* = 7.8, H-9); 8.34 (1H, br. m, NHCO)
7c	1620, 1670, 1685, 1740, 3320	1.74 (3H, s, CH_3); 4.30 (1H, m, H-3); 5.32 (1H, br. m, H-2); 6.22 (1H, d, *J* = 12.3, H-4); 7.32 and 7.36 (2 + 2H, two d, *J* = 7.8, C_6H_4Cl); 7.37-7.48 (2H, m, H-7,8); 7.56 (2H, t, *J* = 7.5, COC_6H_5: H-3,5); 7.63-7.71 (2H, m, H-6 + 4-COC_6H_5: H-4); 7.97 (2H, d, *J* = 7.8, 4-COC_6H_5: H-2,6); 8.19 (1H, d, *J* = 7.8, H-9); 8.39 (1H, br. d, *J* = 7.8, NHCO)
9	1620, 1670, 1740, 3420	2.22 (3H, s, CH3); 4.26 (1H, dd, *J* = 12.3, H-3); 5.15 (1H, d, *J* = 12.6, H-2); 5.49 (1H, m, H-4); 7.23 (1H, t, *J* = 7.5, 3-C_6H_5: H-4); 7.32 (2H, t, *J* = 7.5, 3-C_6H_5: H-3,5); 7.38 (2H, d, *J* = 7.8, 3-C_6H_5: H-2,6); 7.43-7.49 (4H, m, H-7,8 + COC_6H_5: H-3,5); 7.52 (1H, t, *J* = 7.8, COC_6H_5: H-4); 7.63 (2H, d, *J* = 7.2, COC_6H_5: H-2,6); 7.77 (1H, m, H-6); 8.19 (1H, m, H-9); 8.90 (1H, br. d, *J* = 7.5, NHCO, hindered exchange with D2O)
10	1630, 1655, 1680, 1740, 3285, 3450	1.92 (3H, s, CH_3); 4.51 (1H, d, *J* = 7.2, H-3); 5.78 (1H, dd, *J* = 7.2, H-2, changes to a doublet on treatment with D_2O, *J* = 7.5); 7.03-7.05 (2H, m, H-7,8); 7.20-7.30 (4H, 3-C_6H_5: H-2,3,5,6); 7.33 (1H, t, *J* = 7.8, 3-C_6H_5: H-4); 7.46 (2H, t, *J* = 7.5, H-3,5); 7.54-7.57 (2H, m, H-6 + COC_6H_5: H-4); 7.83 (2H, d, *J* = 7.2, COC_6H_5: H-2,6); 7.99 (1H, d, *J* = 7.8, H-9); 8.35 (1H, d, *J* = 7.5, NHCO, disappears on treatment with D_2O); 12.47 (1H, s, H-5, disappears on treatment with D_2O)
11	1630, 1660, 1740, 3270	1.89 (3H, s, 4-$COCH_3$); 1.95 (3H, s, $NCOCH_3$); 7.39 (1H, d, *J* = 7.2, H-3); 5.50 (1H, dd, *J* = 7.5, H-2, changes to a doublet on treatment with D_2O, *J* = 7.2); 6.99-7.02 (2H, m, H-7,8); 7.19-7.34 (5H, m, C_6H_5); 7.55 (1H, d, *J* = 7.8, H-6); 7.96 (1H, d, *J* = 7.8, H-9); 8.03 (1H, d, *J* = 7.2, NHCO, disappears on treatment with D_2O); 12.43 (1H, s, H-5, disappears on treatment with D_2O)

The composition and structure of the synthesised compounds were confirmed by elemental analysis (Table 1) and by IR and ^{1}H NMR spectroscopic data (Table 2).

The position of the oxo group vicinal to the benzimidazole nitrogen atom in compounds **6, 7, 9-11** is evident from the spectroscopic data. The IR spectra show the presence of a carbonyl group absorption band at 1725-1755 cm^{-1} which is at higher frequency than the remaining $\nu_{C=O}$ bands (1660-1690 cm^{-1}) and is characteristic of the compound (see [13]) in which the unshared electron pair of the amide nitrogen atom takes part in the formation of the aromatic conjugated system. In the ^{1}H NMR spectra the H-9 proton is found at lower field (7.96-8.21 ppm) than the remaining aromatic protons since it is deshielded by the sterically nearby carbonyl group.

The presence of a partially hydrogenated pyridine ring in the 4-benzoyl-substituted compounds **6, 7** is confirmed in the ^{1}H NMR spectra by the appearance of the spin spin interaction in the RCONH–CH(C=O)–CH(Ar)–CH(C=O) fragment. The 4-H proton is seen as a clear doublet and the amino group proton as a broadened signal, the doublet nature of which is not clearly seen. The protons corresponding to positions 2 and 3 are seen to higher fields as broadened signals. Such a spectroscopic picture can be explained by restricted conformational interconversion in the tetrahydropyridine ring. On the other hand, in the 4-acetyl analog **9** the multiplicity of the proton signals for position 3 and the amino group are distinctly seen. Evidently in this the conformational mobility of the ring is higher and this can be explained by the increase tendency of the compound to undergo conversion to the prototropic isomer **10**. A deuterium exchange of the amide group proton in compounds **6, 7, 9** is hindered, the intensity of the signal being decreased by 10-20% after 12 h.

In the 1,2,3,5-tetrahydropyrido[1,2-*a*]benzimidazoles **10** and **11** the proton signals are characteristic: H-5 is seen as a singlet with a chemical shift typical of a benzimidazole (see [13]) and H-3 and the amide amino group as a doublet due to interaction with the H-2 proton (the latter being observed as a double doublet). When treated with deuterium oxide the signals for both protons bonded to nitrogen atoms disappear and the H-2 signal becomes a doublet.

In the molecules of the tetrahydro compounds **6, 7, 9-11** the H-2,3 and H-4 protons occupy predominantly axial positions as indicated by their high mutual spin spin coupling values (7.2-12.6 Hz).

Hence cycloaddition of 2-acylmethyl-1H-benzimidazoles to 4-arylidene-1,3-oxadiazol-5-ones provides an efficient method for synthesizing N-(3-aryl-4-acyl-1-oxo-1,2,3,4-tetrahydropyridino[1,2-*a*]benzimidazol-2-yl)benz- and acetamides which, in specific cases, tend to undergo prototropic isomerization to give the rather stable 1,2,3,5-tetrahydropyrido[1,2-*a*]benzimidazoles.

EXPERIMENTAL

Monitoring of the reaction course and the purity of the compounds prepared was carried out by TLC on Silufol UC-254 plates in the solvent system benzene-ethanol (9: 1) and revealed using UV light. IR Spectra were recorded on a UR-20 instrument for KBr tablets. The ^{1}H NMR spectra were taken on a Varian VXR-300 (300 MHz) spectrometer using DMSO-d_6 solvent and TMS internal standard. All of the compounds were dried for 5 h at 110ºC before elemental and spectroscopic investigation.

N-(4-Benzoyl-1-oxo-3-phenyl-1,2,3,4-tetrahydropyrido[1,2-*a*]benzimidazol-2yl)benzamide (6a). A mixture of compound **1a** (0.518 g, 2.2 mmol), azlactone **2a** (0.498 g, 2 mmol), and anhydrous pyridine (2 ml) was held on an oil bath at 120ºC for 1 h. The crystallized reaction mixture was diluted with glacial acetic acid (2 ml) and stirred to a homogenous state. After cooling, the precipitate was filtered off and washed with 2 propanol. The product was obtained in an analytically pure state after drying.

Compounds 6b-f, 7a-c were obtained similarly from compound **1a** and the corresponding azlactones **2b-f, 3a-c**. For the separation of compound **6e** the reaction mixture was mixed with water (2 ml) instead of acetic acid and for **6f** with 2-propanol (2 ml).

N-(4-Acetyl-1-oxo-3-phenyl-1,2,3,4-tetrahydropyrido[1,2-*a*]benzimidazol-2-yl)benzamide (9) and N-(4-Acetyl-1-oxo-3-phenyl-1,2,3,5-tetrahydropyrido[1,2-*a*]benzimidazol-2-yl)benzamide (10). A mixture of compound **1b** (0.174 g, 1 mmol), azlactone **2a** (0.249 g, 1 mmol), and chlorobenzene (1 ml) was heated on an oil bath at 135ºC for 2 h. The reaction mixture was diluted with chlorobenzene (1 ml) and filtered hot. The solid produced was washed with hot chlorobenzene (1 ml) and the filtrate was allowed to cool slowly. The precipitate was recrystallized from a mixture of pyridine and water (1:1) to give compound **9** (0.205 g). A second precipitate was formed on cooling the chlorobenzene solution and this was filtered off and washed with 2-propanol to give analytically pure compound **10** (0.110 g) after drying.

N-(4-Acetyl-1-oxo-3-phenyl-1,2,3,5-tetrahydropyrido[1,2-*a*]benzimidazol-2-yl)acetamide (11). A mixture of compound **1b** (0.174 g, 1 mmol), azlactone **2a** (0.206 g, 1.1 mmol), and chlorobenzene (1 ml) was heated on an oil bath at 135ºC for 2 h. The hot solution was diluted with 2-propanol (1 ml), stirred until crystallization began, and left to cool slowly. The precipitate was filtered off, washed with 2-propanol, and dried to give analytically pure compound **11** (0.213 g).

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