Novel Mono- and Bis(spiro-2-amino-4*H*-pyrans): Alum-Catalyzed Reaction of 4-Hydroxycoumarin and Malononitrile with Isatins, Quinones, or Ninhydrin

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Abstract: Some new mono- and bis(spiro-2-amino-4*H*-pyrans) are synthesized by the multicomponent condensation reaction of 4-hydroxycoumarin and malononitrile with isatins, ninhydrin, 9,10-phenanthrenequinone, or acenaphthenequinone. Alum was found to be a suitable and efficient catalyst for this condensation.

Key words: alum [$KAl(SO_4)_2{\cdot}12H_2O$], spiro-2-amino-4*H*-pyrans, 4-hydroxycoumarin

The 4*H*-pyran derivatives have attracted strong interest due to their useful biological and pharmacological properties such as anticoagulant, spasmolytic, diutretic, anticancer,[1] and antianaphylactin characteristics.[2] 4*H*-Pyrans also occur in various natural products.[3] Development of 4*H*-pyrans synthesis has been of considerable interest in organic synthesis, because of their broad biological and pharmaceutical activities. Spiropyrans are photochromic compounds that have been widely studied because of their potential utility in various high-tech applications.[4]

Oxindole derivatives are known to possess a variety of biological activities, such as potent inhibition of monoamine oxidase in human urine and rat tissues,[5] inhibition of several enzymes such as acetylcholinestrease[6] and atrial natriuretic peptide-stimulated guanylate cyclase, and potent antagonist of in vitro receptor binding by atrial natriuretic peptide,[7] besides possessing a wide range of central nervous system activities.[8] Compounds carrying the indole moiety exhibit antibacterial and antifungal activities.[9] Furthermore, it has been reported that sharing of the indole 3-carbon atom in the formation of spiroindoline derivatives, highly enhances biological activity.[10,11] The derivatives of spirooxindole ring systems are useful as antimicrobial, antitumor agents, and as inhibitors of the human NKI receptor, besides being found in a number of alkaloids like horsifiline, spirotryprostatin, and (+)-elacomine.[12]

Condensed heterocyclic compounds containing a 2-oxindole nucleus or a 4*H*-pyrano fragment have different pharmacological activities, for example, spirocyclic 2-oxindole systems form the basis of such alkaloids as alstonisine and macroxin from the plants of the Alstonia genus[13,14] and bisindole spirocyclic alkaloids from the group of gardmultine, *Gardneria multiflora*,[15] having an antimalarial and ganglioblocking action.

In recent years, the synthesis of spiro compounds via multicomponent condensation reactions has been reported. Many synthetic methodologies have been developed for constructing spirooxindoles, most of which were based on cycloaddition or condensation reactions.[16]

Several methods for the synthesis of spiro-2-amino-4*H*-pyranoxindoles have also been reported and the conventional synthesis involves a three-component condensation of isatin with dimedone or barbituric acid and malononitrile.[17] Thus, each of the known procedures for the synthesis of corresponding spirooxindole-4*H*-pyran system has its merits, however, further studies are still necessary to develop facile, environmental, and economical multicomponent methodology. Due to the biological activity of 4*H*-pyrans containing a 2-oxindole nucleus, and our interest in the synthesis of heterocyclic compounds,[18] we report herein a simple and efficient method for the preparation of new mono- and bis(spiro-2-amino-4*H*-pyran) derivatives using $KAl(SO_4)_2{\cdot}12H_2O$ (alum) as the catalyst in the multicomponent reaction of 4-hydroxycoumarin, malononitrile, and isatins.

Alum is an inexpensive,[19] water-soluble, nontoxic, and commercially available compound that can be used in the laboratory without special precautions. Alum, well known for its traditional role in water treatments (coagulation and clarification) has recently been reported as a reusable, cheap, and excellent catalyst for the preparation of organic compounds, for example, in the transesterification of palm oil,[20] synthesis of α-aminophosphonates,[21] 2,3-dihydroquinazolin-4(1*H*)-ones,[22] 1,3,4-oxadiazoles,[23] dihydropyrimidinones,[24] *cis*-isoquinolonic acids,[19] pyrroles,[25] and coumarins.[26] Thus, continuing our research on new one-pot reactions, we considered alum to be ideal for effecting the synthesis of mono- and bis(spiro-2-amino-4*H*-pyrans) via a three- or pseudo five-component reaction of 4-hydroxycoumarin, malononitrile, and isatins. Some of these compounds have already been prepared in this way by heating in the presence of triethylbenzylammonium chloride.[17c] Herein, we describe our very simple, green, and efficient route to the synthesis of spiro-2-amino-4*H*-pyrans using a catalytic amount of alum in water or ethanol–water media at 25 or 60 °C.

The synthesis of mono-spiro-2-amino-4*H*-pyran derivatives **4a–d** was achieved by the three-component condensation of isatin derivatives **1a–d**, malononitrile **2**, and 4-

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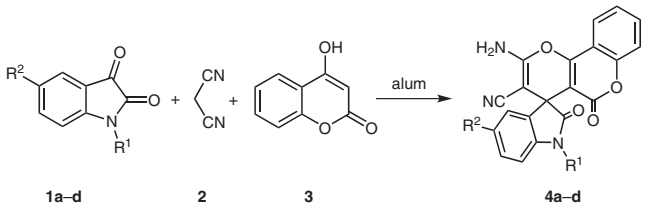
hydroxylcoumarin **3** in the presence of 10 mol% catalyst. The reaction was carried out in water using alum as catalyst at 25 or 60 °C to give products **4a–d** in good to high yields (Scheme 1). Bis(spiro-2-amino-4*H*-pyran) derivatives **6a**,**b** were obtained by pseudo five-component reaction of bis-isatins **5a**,**b** (1 mmol), malononitrile **2** (2 mmol), and 4-hydroxycoumarin **3** (2 mmol) in aqueous ethanol (Scheme 2).



Scheme 1 Three-component synthesis of spiro-2-amino-4*H*-pyrans **4a–d**. *Reagents and conditions*: H_2O, r.t., 15 h or 60 °C, 5 h.

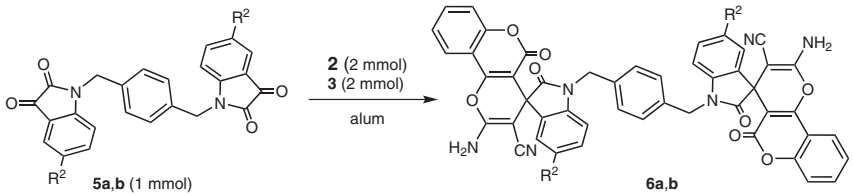
In order to optimize the conditions, we used isatin (**1a**), malononitrile (**2**), and 4-hydroxycoumarin (**3**) and tested various amounts of alum as catalyst. After two hours with 3, 6, 10, and 15 mol% of alum, yields of 65, 83, 93, and 89%, respectively, were obtained (Table 1, entries 1–4). In the absence of alum the product was obtained in low yield of (Table 1, entry 5).

It was found that room temperature was appropriate for the alum-catalyzed reaction (Table 1, entry 6). The high temperature could improve the reaction rate and shorten the reaction time (Table 1, entry 3). The solvents also played an important role in this reaction catalyzed by alum (Table 1, entries 3 and 7–11). Several solvents were tested for the reaction, such as THF, MeCN, DMSO, DMF, EtOH–H_2O, and H_2O. The reaction hardly proceeded in THF. However, the reaction in H_2O afforded product in high yield with nearly complete conversion. Therefore, water was selected as the reaction solvent in the following investigation (Table 1, entry 3).

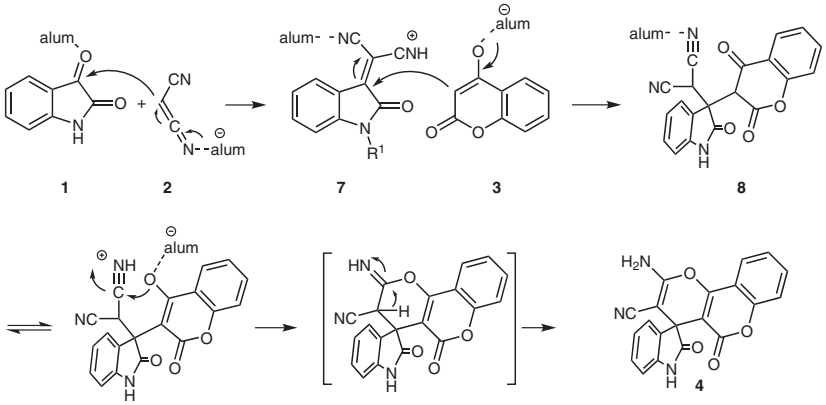
Table 1 Optimization of Reaction for the Synthesis of **4a**

Entry	Catalyst[a]	Solvent	Temp (°C)	Yield (%)[b]
1	alum (3 mol%)	H_2O	60	65
2	alum (6 mol%)	H_2O	60	83
3	alum (10 mol%)	H_2O	60	93
4	alum (15 mol%)	H_2O	60	89
5	–	H_2O	60	27
6	alum (10 mol%)	H_2O	r.t.[c]	85
7	alum (10 mol%)	EtOH–H_2O[d]	60	65
8	alum (10 mol%)	MeCN	60	50
9	alum (10 mol%)	DMF	60	55
10	alum (10 mol%)	THF	60	20
11	alum (10 mol%)	DMSO	60	40

[a] Reaction time = 2 h.
[b] Yields refer to isolated products.
[c] Reaction time = 15 h.
[d] EtOH–H_2O (5:2).



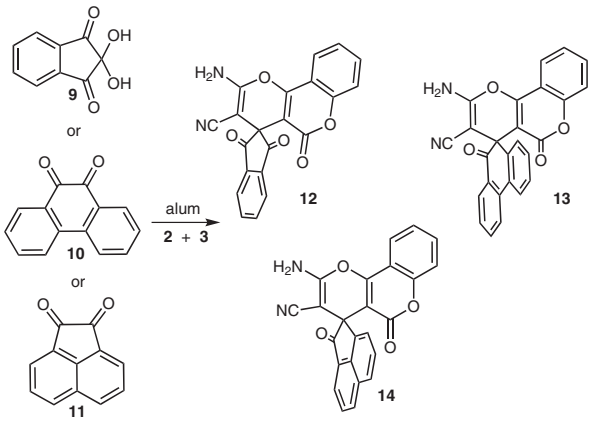
Scheme 2 Pseudo five-component synthesis of bis(spiro-2-amino-4*H*-pyrans) **6a**,**b**. *Reagents and conditions*: EtOH–H_2O (5:2), r.t., 24 h or 60 °C, 5 h.



Scheme 3 Plausible mechanism of the alum-catalyzed synthesis of spiro-2-amino-4*H*-pyrans

Although we have not yet established the mechanism, a possible explanation is given in Scheme 3. The process represents a typical cascade reaction in which the isatin (**1**) first condenses with malononitrile (**2**) to afford isatylidene malononitrile derivative **7** in the presence of alum in water. This step was regarded as a fast Knoevenagel condensation. Then, **7** is attacked via Michael addition of 4-hydroxycoumarin (**3**) to give the intermediate **8** followed by the cycloaddition of hydroxyl group to the cyano moiety to form the desired product **4** (Scheme 3).

Also this approach can be used for the synthesis of spiro-2-amino-4*H*-pyrans **12**, **13**, and **14** when ninhydrin (**9**), 9,10-phenanthroquinone (**10**), or acenaphthenequinone (**11**) were used as starting materials (Scheme 4).



Scheme 4 Synthesis of spiro-2-amino-4*H*-pyrans **12–14**

The mono- and bis(spiro-2-amino-4*H*-pyrans) synthesized by our method are listed in Table 2. Apparently, recycling of catalyst is possible for four successive times without significant loss of activity (Table 2, **4a**). All products were characterized by melting point, IR, ^{1}H NMR, and ^{13}C NMR spectral data, as well as by elemental analyses.

In conclusion, we have described an efficient, one-pot, and three- or pseudo five-component method for the synthesis of mono- and bis(spiro-2-amino-4*H*-pyrans) catalyzed by alum. Short reaction times, high yields, and easy workup are the advantages of this protocol.

Mps were measured on an Electrothermal 9100 apparatus. IR spectra were measured on a Unicom Galaxy Series FTIR 5000 spectrophotometer. ^{1}H NMR and ^{13}C NMR spectra were determined on a Bruker Avance 300 MHz spectrometer. Elemental analyses were performed using a Vario EL III elemental analyzer.

1-Benzylisatin (1c)

Isatin (**1a**; 147 mg, 1 mmol) and benzyl chloride (252 mg, 2 mmol) were stirred in anhyd DMSO (2 mL) in the presence of K_2CO_3 (165 mg, 1.2 mmol) at 25 °C for 20 h. Compound **1c** was precipitated by addition of H_2O and collected by filtration and recrystallized from EtOH; yield: 184 mg (78%).

Table 2 Alum-Catalyzed Synthesis of Mono- and Bis(spiro-2-amino-4*H*-pyrans)

Product	R^1	R^2	Time (h)[a]	Yield (%)[a,b]	Time (h)[c]	Yield (%)[b,c]	Mp (°C)	Lit.[17c] mp (°C)
4a[d]	H	H	2	93 (90, 87, 83, 80)[e]	15	86	303	292–294
4b[d]	H	Br	2	91	15	82	336	>300
4c[d]	Bn	H	2.5	88	18	76	278	277–279
4d[d]	Me	H	2.5	89	18	77	286	287–288
6a[f]	–	H	5	74	24	60	318	–
6b[f]	–	Br	5	69	24	58	268	–
12[d]	–	–	2	92	10	85	246	–
13[f]	–	–	5	65	24	54	285	–
14[f]	–	–	5	63	24	53	257	–

[a] At 60 °C.
[b] Yields refer to isolated products.
[c] At r.t.
[d] Reaction solvent: H_2O.
[e] Yields after recycling of catalyst.
[f] Reaction solvent: EtOH–H_2O (5:2).

Bis-isatins 5a,b

Isatin (**1a**; 147 mg, 1 mmol) or 5-bromoisatin (**1b**; 225 mg, 1 mmol) and α,α′-dibromo-*p*-xylene (263 mg, 0.5 mmol) were stirred in anhyd DMSO (2 mL) in the presence of K_2CO_3 (165 mg, 1.2 mmol) at r.t. for 24 h. Compound **5a** or **5b** was precipitated by the addition of H_2O and collected by filtration and recrystallized from EtOH; yield: **5a**: 265 mg (67%); **5b**: 332 mg (60%).

Mono- and Bis(spiro-2-amino-4*H*-pyrans) 4a–d and 6a,b; General Procedure

A mixture of isatin **1a–d** or **5a**,**b** (1 mmol), malononitrile (**2**; 1 mmol for **1a–c** and 2 mmol for **5a**,**b**), 4-hydroxycoumarin (**3**; 1 mmol for **1a–c** and 2 mmol for **5a**,**b**), and alum (10 mol%, 50 mg) in H_2O or aq EtOH (5 mL) was stirred at r.t. or 60 °C for several hours (Table 2). Upon completion of the reaction as monitored by TLC (*n*-hexane–EtOAc, 2:1), the reaction mixture was allowed to cool to r.t. The solid was collected by filtration and washed with H_2O (15 mL) and cold EtOH (15 mL) to give the desired products. The crude products were recrystallized from EtOH to yield mono- and bis(spiro-2-amino-4*H*-pyrans) **4a–d** and **6a**,**b**.

4a

IR (KBr): 3397, 3296, 3196, 2206, 1710, 1674, 1604, 1359 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): δ = 6.84–6.93 (m, 2 H_{arom}), 7.16–7.23 (m, 2 H_{arom}), 7.49–7.58 (m, 2 H_{arom}), 7.69 (s, 2 H, NH_2), 7.75–7.80 (t, *J* = 7.38 Hz, 1 H_{arom}), 7.93 (d, *J* = 7.92 Hz, 1 H_{arom}), 10.70 (s, 1 H, NH).

4b

IR (KBr): 3429, 3325, 3207, 2200, 1710, 1998, 1606, 1359 cm^{-1}.

^{1}H NMR (300 MHz, DMSO-d_6): δ = 6.81 (d, *J* = 8.25 Hz, 1 H_{arom}), 7.38–7.41 (dd, *J* = 8.25, 2.05 Hz, 1 H_{arom}), 7.49–7.57 (m, 1 H_{arom}), 7.55 (s, 2 H, NH_2), 7.75–7.80 (m, 3 H_{arom}), 7.92–7.95 (dd, *J* = 7.90, 1.40 Hz, 1 H_{arom}), 10.82 (s, 1 H, NH).

4c

IR (KBr): 3429, 3344, 3180, 2204, 1716, 1672, 1606, 1361 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 4.96–4.98 (ABq, 2 H, CH_2), 6.79 (t, J = 7.70 Hz, 1 H_{arom}), 7.00–7.02 (t, J = 7.42 Hz, 1 H_{arom}), 7.18–7.34 (m, 5 H_{arom}), 7.46–7.56 (m, 4 H_{arom}), 7.76–7.81 (m, 2 H_{arom}), 7.79 (s, 2 H, NH_2), 7.96–7.99 (dd, J = 7.90, 1.41 Hz, 1 H_{arom}).

4d

IR (KBr): 3444, 3304, 3170, 2208, 1713, 1675, 1606, 1361 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 3.24 (s, 3 H, CH_3), 7.05–7.13 (m, 2 H_{arom}), 7.32–7.40 (m, 2 H_{arom}), 7.53–7.62 (m, 2 H_{arom}), 7.75–7.80 (m, 3 H, NH_2 and 1 H_{arom}), 7.97 (d, ^3J = 7.85 Hz, 1 H_{arom}).

6a

IR (KBr): 3405, 3308, 3173, 2198, 1720, 1676, 1608, 1356 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 4.95 (ABq, 4 H, CH_2), 6.79–6.82 (d, J = 7.90 Hz, 2 H_{arom}), 6.98–7.02 (t, J = 6.70 Hz, 2 H_{arom}), 7.19–7.24 (t, J = 8.20 Hz, 3 H_{arom}), 7.31–7.34 (d, J = 7.39 Hz, 2 H_{arom}), 7.43 (s, 4 H, NH_2), 7.51–7.59 (m, 4 H_{arom}), 7.78 (s, 5 H_{arom}), 7.96–7.99 (d, J = 8.03 Hz, 2 H_{arom}).

^{13}C NMR (75 MHz, DMSO-d_6): δ = 43.70, 47.82, 57.17, 101.58, 109.67, 112.88, 117.168, 117.52, 123.26, 123.45, 124.52, 125.55, 127.68, 129.52, 132.72, 133.14, 134.28, 135.36, 143.17, 152.53, 155.81, 158.88, 159.09, 176.42.

Anal. Calcd for $C_{48}H_{28}N_6O_8$: C, 70.58; H, 3.46; N, 10.29. Found: C, 71.08; H, 3.28; N, 10.78.

6b

IR (KBr): 3427, 3342, 3115, 2198, 1716, 1674, 1606, 1359 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 4.96 (ABq, 4 H, CH_2), 6.76 (s, 2 H_{arom}), 7.09–7.15 (m, 3 H_{arom}), 7.41 (s, 4 H, NH_2), 7.55–7.96 (m, 13 H_{arom}).

^{13}C NMR (75 MHz, DMSO-d_6): δ = 43.71, 47.94, 56.54, 100.92, 111.59, 113.07, 115.36, 117.19, 117.46, 123.30, 125.52, 127.70, 132.21, 133.21, 134.29, 135.02,136.34, 137.64, 142.46, 152.60, 156.15, 159.07, 159.19, 176.08.

Anal. Calcd for $C_{48}H_{26}Br_2N_6O_8$: C, 59.16; H, 2.69; N, 8.62. Found: C, 58.48; H, 2.83; N, 8.33.

Spiro-2-amino-4*H*-pyrans 12, 13, and 14

The spiro-2-amino-4*H*-pyrans **12**, **13**, and **14** were prepared as described for the above products by three-component reaction of ninhydrin (**9**; 1 mmol), 9,10-phenanthroquinone (**10**, 1 mmol), or acenaphthenequinone (**11**, 1 mmol) with **2** (1 mmol) and **3** (1 mmol).

12

IR (KBr): 3394, 3321, 3189, 1714, 1672, 1359 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 7.55–7.62 (m, 3 H_{arom}), 7.80–7.88 (m, 2 H_{arom}), 7.92–7.97 (m, 2 H_{arom}), 8.06 (s, 1 H_{arom}), 8.13 (s, 2 H, NH_2).

^{13}C NMR (75 MHz DMSO-d_6): δ = 52.74, 53.21, 100.16, 112.37, 116.94, 117.32, 117.49, 123.00, 124.12, 125.53, 125.91, 126.79, 134.75, 135.68, 137.83, 140.94, 152.66, 157.20, 160.04, 160.22, 199.30.

13

IR (KBr): 3400, 3321, 2212, 1737, 1606, 1541, 1413 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 7.37–7.97 (m, 12 H_{arom}), 8.86 (s, 2 H, NH_2).

Anal. Calcd for $C_{26}H_{14}N_2O_4$: C, 74.64; H, 3.37; N, 6.70. Found: C, 74.29; H, 3.46; N, 6.62.

14

IR (KBr): 3450, 3319, 2222, 1728, 1637, 1579 cm^{-1}.

^1H NMR (300 MHz, DMSO-d_6): δ = 7.48–8.37 (m, 12 H, NH_2 and 10 H_{arom}).

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