Microwave-assisted Aqueous Multicomponent Reaction: Facile Synthesis of Polyfunctionalized Indoline-spiro Fused Pyran Derivatives

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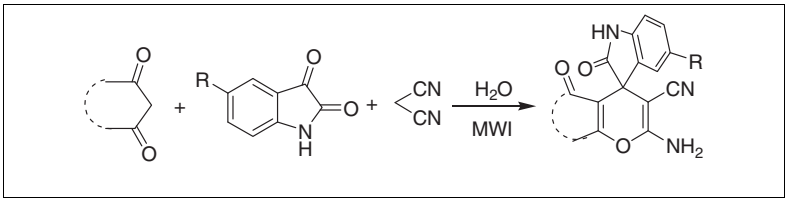

A green and highly efficient method for the synthesis of polyfunctionalized indoline-spiro fused pyran derivatives has been established. This reaction was conducted by reacting readily available and inexpensive starting materials, such as isatins, cyclic-1,3-dicarbonyl compounds, and malononitrile in aqueous media without any catalysts under microwave irradiation. The present green synthesis shows fascinating properties such as the use of water as the reaction solution, concise one-pot conditions, short reaction periods (8–14 min), and easy purifications. The synthesis could also set a good example to GAP (Group-Assistant-Purification) chemistry in which purification via chromatography can be avoided and the pure products can be easily acquired only by washing the crude products with 95% EtOH.

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INTRODUCTION

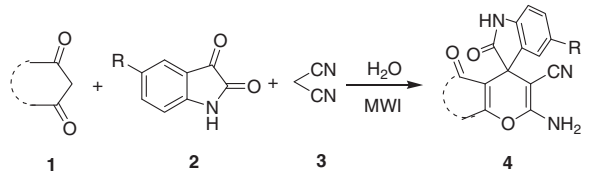
The need to reduce the amount of toxic waste and by-product arising from chemical process requires increasing emphasis on the use of less toxic and environmentally compatible materials in the design of new synthetic methods. One of the most promising approaches is using water as reaction media. There has been growing recognition that water is an attractive medium for many organic reactions [1]. Multicomponent reactions (MCRs), especially those performed in aqueous medium, have enjoyed a very high prestige in the present synthesis of chemically and biologically important compounds on account of their environmentally friendly atom-economy and green characteristics [2–4]. These reactions provide a wide range of possibilities for the efficient construction of a variety of invaluable products in a single operation instead of tedious multi-step synthesis, thus avoiding complicated purifications and saving both solvents and reagents [5]. Therefore, one-pot MCRs can dramatically reduce the generation of chemical waste and the cost of the starting materials, shorten reaction periods, giving higher overall chemical yields [6].

The spirooxindole derivatives are attractive targets in organic synthesis by virtue of their highly evidently biological activities as well as wide-ranging utility as synthetic intermediates for alkaloids, drug candidates, and clinical pharmaceuticals [7]. Furthermore, it has been reported that sharing of the indole 3-carbon atom in the formation of spiroindoline derivatives can highly enhance biological activity [8–10]. Therefore, the synthesis of these compounds has attracted great attentions [11]. As a result, there have been a few reports [12] about the aqueous MCRs for the synthesis of spirooxindole derivatives, and different catalysts, such as L-proline [12], TEBA [12], and NH_4Cl [12], were used in these reactions, increasing the complexity of purifications. As a consequence of our interest in aqueous synthesis [13] and our continual work on the fast synthesis of heterocycles [14], we investigated a high-efficient green approach to spirooxindoles performed by reacting isatins, malononitrile, and cyclic-1,3-dicarbonyl substrates as starting materials in water under microwave irradiation without any catalysts (Scheme 1).

RESULTS AND DISCUSSION

We began this study by subjecting tetronic acid and 5-fluoroindole-2,3-dione to react with malononitrile in various solvents such as DMF, TFA, Glycol, EtOH, AcOH, and water under microwave irradiation for condition optimization (Scheme 2). Although 2-amino-5′-fluoro-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile can be achieved in the presence of the given solvents except DMF and TFA, HOAc and water resulted in higher chemical yield, and the pure water as solvent can achieve a very high yield of 90%, even 2% higher than that of HOAc when the reaction was performed at 80°C for 10 min under microwave irradiation, as a good absorber for microwave

Scheme 1



Scheme 2

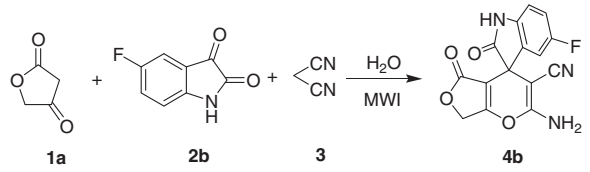


Table 1

Optimization for the synthesis of **4b** under MW.

Entry	Solvent	T/°C	Time (min)	Yield (%)
1	DMF	80	35	trace
2	TFA	80	40	trace
3	Glycol	80	25	45
4	EtOH	80	15	70
5	HOAc	80	10	88
6	H_2O	80	10	90
7	H_2O	60	20	80
8	H_2O	90	10	89
9	H_2O	100	10	87

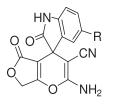
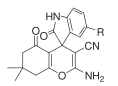
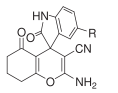
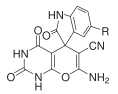
energy in view of its relatively environmental-friendly characteristics (Table 1) [15].

To further optimize the reaction temperature, the reaction was carried out at different temperature ranging from 60 to 100°C. We found that the yield of product **4** was improved, and the reaction time was shortened as the reaction temperature was increased to 80°C. No significant improvement in yield was obtained past that point, so 80°C was chosen as the reaction temperature for all further studies.

Under these optimized conditions, the reaction scope was evaluated by using various isatins and 1,3-dicarbonyl substrates. Commercially available indoline-2,3-dione bearing either electron-withdrawing or electron-donating functional groups such as fluoro, chloro, bromo, or methyl were all found to be suitable for the reaction with tetronic acid and malononitrile to obtain spiro[indoline-3′4-pyran]-2-one derivatives in very good yields of 89–92% under microwave heating (Table 2, entries 1–4). We also utilized 5,5-dimethylcyclohexane-1,3-dione, cyclohexane-1,3-dione, and dihydro-2,2-dimethylpyrimidine-4,6(1H,5H)-dione instead of tetronic acid to afford the corresponding spiro[indoline-3′4-pyran]-2-ones within 8–14 min in very good yields (87–95%)(Table 2, entries 5–12). After careful analysis, we are also delighted to observe the delicate electronic effects: isatins with electron-withdrawing groups reacted much rapidly, whereas electron-rich groups decreased the reactivity, requiring longer reaction times.

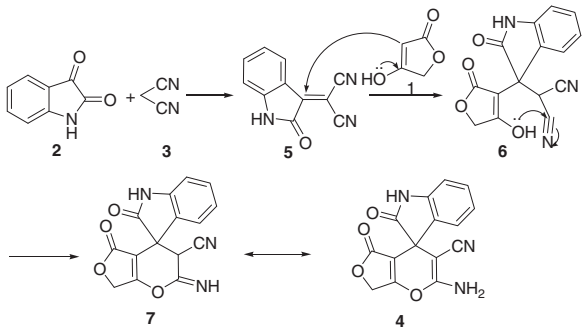
Table 2

Synthesis of compound **4** in water under microwave heating.

Entry	Product		R	Time (min)	Yield (%)
1		**4a**	Me	12	89
2		**4b**	F	10	90
3	**4a–4d**	**4c**	Cl	8	92
4		**4d**	Br	8	90
5		**4e**	H	12	87
6	**4e–4h**	**4f**	F	8	95
7		**4g**	Cl	9	91
8		**4h**	Br	9	90
9	**4i–4j**	**4i**	Me	14	88
10		**4j**	H	9	90
11		**4k**	F	8	93
12	**4k–4l**	**4l**	Cl	9	91

Given the large number of commercially available cyclic-1,3-dicarbonyl substrates and the easy access to isatins, the present method should be applied in the synthesis of libraries with high functional group diversity. We expect this method to find application in the field of combinatorial chemistry, diversity-oriented synthesis, and drug discovery. In this study, all the products were characterized by melting point, IR, and ^{1}H NMR spectral data as well as HRMS (ESI).

Scheme 3



Plausible mechanism for the synthesis of 2-amino-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile **4** was described in Scheme 3. The process represents a typical cascade reaction in which the isatin **2** first condenses with malononitrile **3** to afford isatylidene malononitrile derivative **5** in water. This step was regarded as a fast Knoevenagel condensation. Then, **5** is attacked via Michael addition of tetronic acid **1** to give the intermediate **6** followed by the cycloaddition of hydroxyl group to the cyano moiety to form the desired product **4**.

In summary, a green and high-efficient approach to spirooxindole derivatives has been developed by performing the reaction in one-pot fashion in water under microwave irradiation without any catalyst. The reactions showed a broad substrates of using readily available and inexpensive starting materials of various isatins, malononitrile, and cyclic-1,3-dicarbonyl compounds. Particularly, this green synthesis shows several attractive characteristics such as the use of water as the reaction medium, simple conditions, short reaction periods, easy work-up, straightforwardness of the procedure, and reduced waste production by the lack of any requirement for catalysts.

EXPERIMENTAL

Microwave irradiation was carried out with Initiator from Biotage Company, Uppsala, Sweden. Melting points were determined in open capillaries and were uncorrected. IR spectra were recorded on a FTIR-Tensor 27 spectrometer (Bruker Company, Ettlingen, Germany). ^{1}H NMR spectra were measured on a Bruker DPX 400 (100) MHz spectrometer (Ettlingen, Germany) in DMSO-d_6 with chemical shift (δ) given in ppm relative to TMS as internal standard. The exact mass measurements were obtained by high resolution mass instrument (GCT-TOF instrument).

General procedure for the synthesis of compounds 4 under MW. Typically, in a 10-mL reaction vial, isatins (1 mmol), malononitrile (1 mmol), cyclic-1,3-dicarbonyl compounds (1 mmol), and water (2 mL) were mixed and then with 20-s pre-stiring before being capped. The mixture was heated for a given time at 80°C under microwave irradiation. Upon completion (as monitored by TLC), the reaction mixture was cooled to room temperature and then filtered to give the crude product, which was further washed by the 95% EtOH to give the pure product **4**.

2-Amino-5′-methyl-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile (4a). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.59 (s, 1H, NH), 7.67 (s, 2H, NH_2), 7.05 (d, J = 8.0 Hz, 1H, ArH), 7.01 (s, 1H, ArH), 6.75 (d, J = 7.6 Hz, 1H, ArH), 5.19 (d, J = 16 Hz, 1H, CH_2), 5.09 (d, J = 17.2 Hz, 1H, CH_2), 2.24 (s, 3H, CH_3); IR (KBr, ν, cm^{-1}): 3333, 3200, 2913, 2203, 1776, 1717, 1691, 1597, 1495, 1374, 1340, 1258, 1101, 1027, 1002, 931, 821, 765, 690, 596; HRMS (ESI): *m/z* calcd for: $C_{16}H_{11}N_3NaO_4$, 332.0642, found: 332.0645.

2-Amino-5′-fluoro-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile (4b). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.74 (s, 1H, NH), 7.76 (s, 2H, NH_2), 7.21 (d, J = 8.0 Hz, 1H, ArH), 7.12–7.07 (m, 1H, ArH), 6.88–6.85 (m, 1H, ArH), 5.21 (d, J = 16.8 Hz, 1H, CH_2), 5.05 (d, J = 16.8 Hz, 1H, CH_2); IR (KBr, ν, cm^{-1}): 3334, 3206, 3089, 2203, 1777, 1719, 1693, 1632, 1594, 1492, 1377, 1342, 1255, 1051, 1029, 1000, 828, 802, 763, 689, 632, 594; HRMS (ESI): *m/z* calcd for: $C_{15}H_8FN_3NaO_4$, 336.0392, found: 336.0384.

2-Amino-5′-chloro-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile (4c). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.85 (s, 1H, NH), 7.78 (s, 2H, NH_2), 7.38 (s, 1H, ArH), 7.31 (d, J = 8.4 Hz, 1H, ArH), 6.89 (d, J = 8.0 Hz, 1H, ArH), 5.21 (d, J = 16.8 Hz, 1H, CH_2), 5.04 (d, J = 16.4 Hz, 1H, CH_2); IR (KBr, ν, cm^{-1}): 3335, 3201, 3092, 2203, 1778, 1719, 1696, 1631, 1597, 1480, 1375, 1341, 1315, 1047, 1028, 1003, 824, 771, 718, 687, 632, 557; HRMS (ESI): *m/z* calcd for: $C_{15}H_8ClN_3NaO_4$, 352.0096, found: 352.0096.

2-Amino-5′-bromo-2′,5-dioxo-5,7-dihydrospiro[furo[3,4-b]pyran-4,3′-indoline]-3-carbonitrile (4d). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.86 (s, 1H, NH), 7.79 (s, 2H, NH_2), 7.49 (s, 1H, ArH), 7.44 (d, J = 8.4 Hz, 1H, ArH), 6.84 (d, J = 8.4 Hz, 1H, ArH), 5.20 (d, J = 16.8 Hz, 1H, CH_2), 5.04 (d, J = 16.8 Hz, 1H, CH_2); IR (KBr, ν, cm^{-1}): 3338, 3184, 3046, 2202, 1773, 1720, 1698, 1634, 1598, 1478, 1373, 1340, 1316, 1253, 1106, 1027, 1003, 815, 771, 631, 542; HRMS (ESI): *m/z* calcd for: $C_{15}H_8BrN_3NaO_4$, 395.9591, found: 395.9591.

2-Amino-7,7-dimethyl-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4e). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.39 (s, 1H, NH), 7.22 (s, 2H, NH_2), 7.14 (t, J = 7.6 Hz, 1H, ArH), 6.97 (d, J = 7.2 Hz, 1H, ArH), 6.89 (t, J = 7.2 Hz, 1H, ArH), 6.79 (d, J = 7.6 Hz, 1H, ArH), 2.56 (s, 2H, CH_2), 2.13 (dd, J_1 = 16 Hz, J_2 = 15.6 Hz, 2H, CH_2), 1.02 (d, J = 12.4 Hz, 6H, CH_3); IR (KBr, ν, cm^{-1}): 3377, 3314, 3143, 2959, 2927, 2192, 1722, 1682, 1655, 1620, 1604, 1471, 1349, 1327, 1298, 1223, 1166, 1055, 903, 746, 679, 615, 559; HRMS (ESI): *m/z* calcd for: $C_{19}H_{17}N_3NaO_3$, 358.1163, found: 358.1172.

2-Amino-5′-fluoro-7,7-dimethyl-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4f). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.41 (s, 1H, NH), 7.28 (s, 2H, NH_2), 6.99–6.94 (m, 2H, ArH), 6.79–6.66 (m, 1H, ArH), 2.55 (s, 2H,CH_2), 2.15 (s, 2H, CH_2), 1.02 (d, J = 4.0 Hz, 6H, CH_3); IR (KBr, ν, cm^{-1}): 3381, 3304, 3158, 2959, 2932, 2191, 1725, 1680, 1654, 1599, 1487, 1459, 1351, 1329, 1299, 1269, 1184, 1058, 880, 810, 794, 620, 597; HRMS (ESI): *m/z* calcd for: $C_{19}H_{16}FN_3NaO_3$, 376.1068, found: 376.1068.

2-Amino-5′-chloro-7,7-dimethyl-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4g). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.54 (s, 1H, NH), 7.32 (s, 2H, NH_2), 7.21–7.18 (m, 1H, ArH), 7.10 (s, 1H, ArH), 6.80 (d, J = 8.4 Hz, 1H, ArH), 2.62–2.54 (m, 2H, CH_2), 2.16 (s, 2H, CH_2), 1.02 (s, 6H, CH_3); IR (KBr, ν, cm^{-1}): 3371, 3289, 3155, 2958, 2930, 2193, 1727, 1681, 1653, 1603, 1478, 1350, 1329, 1225, 1167, 1058, 887, 811, 637, 619, 560; HRMS (ESI): *m/z* calcd for: $C_{19}H_{16}ClN_3NaO_3$, 392.0773, found: 392.0793.

2-Amino-5′-bromo-7,7-dimethyl-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4h). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.56 (s, 1H, NH), 7.33 (s, 2H, NH_2), 7.32–7.31 (m, 1H, ArH), 7.21 (s, 1H, ArH), 6.76 (d, J = 8.0 Hz, 1H, ArH), 2.62–2.53 (m, 2H, CH_2), 2.16 (dd, J_1 = 16.4 Hz, J_2 = 3.2 Hz, 2H, CH_2), 1.02 (s, 6H, CH_3); IR (KBr, ν, cm^{-1}): 3367, 3290, 3159, 2958, 2929, 2194, 1727, 1681, 1655, 1604, 1475, 1350, 1328, 1224, 1168, 1057, 874, 809, 688, 636, 620; HRMS (ESI): *m/z* calcd for: $C_{19}H_{16}BrN_3NaO_3$, 436.0268, found: 436.0268.

2-Amino-5′-methyl-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4i). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.28 (s, 1H, NH), 7.19 (s, 2H, NH_2),

6.94 (d, $J = 8.0$ Hz, 1H, ArH), 6.81 (s, 1H, ArH), 6.66 (d, $J = 7.6$ Hz, 1H, ArH), 2.65 (s, 2H, CH_2), 2.23 (s, 2H, CH_2), 2.21 (s, 3H, CH_3), 1.93(m, 2H, CH_2); IR (KBr, ν, cm^{-1}): 3361, 3143, 2950, 2195, 1711, 1681, 1658, 1603, 1491, 1351, 1334, 1315, 1213, 1195, 1075, 1008, 955, 830, 689, 649, 542; HRMS (ESI): *m/z* calcd for: $C_{18}H_{15}N_3NaO_3$, 344.1006, found: 344.1025.

2-Amino-2′,5-dioxo-5,6,7,8-tetrahydrospiro[chromene-4,3′-indoline]-3-carbonitrile (4j). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 10.41 (s, 1H, NH), 7.24 (s, 2H, NH_2), 7.14 (t, $J = 7.2$ Hz, 1H, ArH), 7.00 (d, $J = 7.2$ Hz, 1H, ArH), 6.88 (t, $J = 7.2$ Hz, 1H, ArH), 6.77 (d, $J = 7.6$ Hz, 1H, ArH), 2.66 (t, $J = 6.0$ Hz, 2H, CH_2), 2.25–2.20 (m, 2H, CH_2), 1.95–1.91 (m, 2H, CH_2); IR (KBr, ν, cm^{-1}): 3368, 3300, 3161, 2955, 2196, 1715, 1683, 1657, 1608, 1466, 1353, 1335, 1314, 1219, 1197, 1078, 1011, 737, 681, 659, 590; HRMS (ESI): *m/z* calcd for: $C_{17}H_{13}N_3NaO_3$, 330.0850, found: 330.0855.

7′-Amino-5-fluoro-2,2′,4′-trioxo-1′,2′,3′,4′-tetrahydrospiro[indoline-3,5′-pyrano[2,3-d]pyrimidine]-6′-carbonitrile (4k). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 12.32 (s, 1H, NH), 11.15 (s, 1H, NH), 10.50 (s, 1H, NH), 7.43 (s, 2H, NH_2), 7.16 (d, $J = 7.6$ Hz, 1H, ArH), 6.99 (t, $J = 8.8$ Hz, 1H, ArH), 6.78–6.76 (m, 1H, ArH); IR (KBr, ν, cm^{-1}): 3404, 3328, 3216, 3059, 2758, 2207, 1671, 1530, 1487, 1391, 1332, 1188, 1112, 1005, 802, 689, 666, 600, 555; HRMS (ESI): *m/z* calcd for: $C_{15}H_8FN_5NaO_4$, 364.0453, found: 364.0453.

7′-Amino-5-chloro-2,2′,4′-trioxo-1′,2′,3′,4′-tetrahydrospiro[indoline-3,5′-pyrano[2,3-d]pyrimidine]-6′-carbonitrile (4l). mp: >300°C; ^{1}H NMR (400 MHz, DMSO-d_6) (δ, ppm): 12.32 (s, 1H, NH), 11.15 (s, 1H, NH), 10.61 (s, 1H, NH), 7.44 (s, 2H, NH_2), 7.34 (s, 1H, ArH), 7.21 (d, $J = 8.0$ Hz, 1H, ArH), 6.80 (d, $J = 8.4$ Hz, 1H, ArH), IR (KBr, ν, cm^{-1}): 3402, 3322, 3213, 3052, 2819, 2200, 1693, 1645, 1541, 1477, 1406, 1342, 1166, 1128, 1000, 814, 576; HRMS (ESI): *m/z* calcd for: $C_{15}H_8FN_5NaO_4$, 380.0158, found: 380.0158.

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