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ONE-POT, THREE-COMPONENT, GREEN SYNTHESIS OF SOME INDENO[2`,3`:5,6] PYRIDO[2,1-*b*]BENZOTHIAZOLES AND INDENO[2`,3`-*e*]THIAZOLO[3,2 *a*]PYRIDINES

Essam M. Hussein

Department of Chemistry, Faculty of Science, Assiut University, Assiut 71516, Egypt
e-mail: essam.hussein78@yahoo.com

Abstract
The green synthesis of some indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**) via a one-pot, three-component reaction of indan-1,3-dion (**1**), aromatic aldehydes (**2a-f**), (benzothiazol-2-yl)acetonitrile (**3**) or 2-cyanomethyl-4-thiazolinone (**5**), and catalyzed by *p*-toluene sulfonic acid (*p*-TSA) using water as reaction medium is reported.

Keywords: Green synthesis, (benzothiazol-2-yl)acetonitrile, 2-cyanomethyl-4-thiazolinone, pyrido[2,1-*b*]benzothiazole, thiazolo[3,2-*a*]pyridines

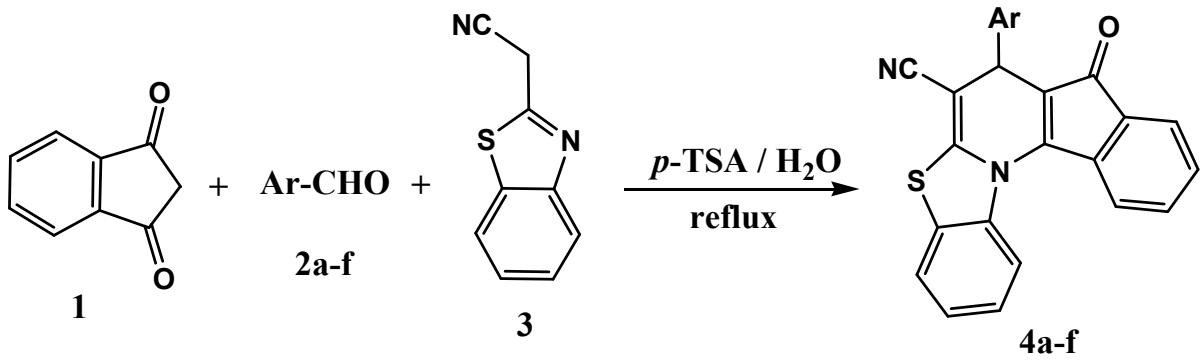
Introduction

The development of organic reactions in water has become highly desirable in recent years to meet environmental considerations.[1] The use of water as a sole medium for organic reactions would seriously contribute to the extension of environmentally benign processes. In this context, in recent years, much attention has been focused on organic reactions in water. Multi-component reactions (MCRs), due to their productivity, simple procedures, and facile execution, are one of the best tools in synthetic chemistry.[2] therefore, the design of novel multi-component reactions has attracted great attention from research groups working in areas such as materials science, drug design, and organic synthesis. As a result, the number of new MCRs in water has grown rapidly.[3-6] Our search of the literature revealed that, some thiazolo[3,2–*a*]pyridine derivatives have been reported to possess antibacterial[7], bactericide[8], coronary dilator, antihypertensive and muscle relaxant activities.[9] As part of our continuing efforts on the development of new routes for the synthesis of biologically active heterocycles,[10-17] Herein, I report a simple and green synthesis of novel indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**) via a one-pot, three-component reaction of indan-1,3-dion (**1**), aromatic aldehydes (**2a-f**), (benzothiazol-2-yl)acetonitrile (**3**)[18] or 2-cyanomethyl-4-thiazolinone (**5**)[19], catalyzed by *p*-toluene sulfonic acid (*p*-TSA) using water as reaction medium

Results and Discussion

The one-pot**,** three-component reaction of indan-1,3-dione (**1**), various aromatic aldehyde (**2a-f**) and (benzothiazol-2-yl)acetonitrile (**3**) proceeded smoothly in refluxing water in the presence of catalytic amount of *p*-toluene sulfonic acid (*p*-TSA) and were complete after short to moderate

reaction time (3-7 h) to afford 7-aryl-6-cyanoindeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-ones (**4a-f**) in good yield (75-89%) (**Scheme 1**, **Table 1**).

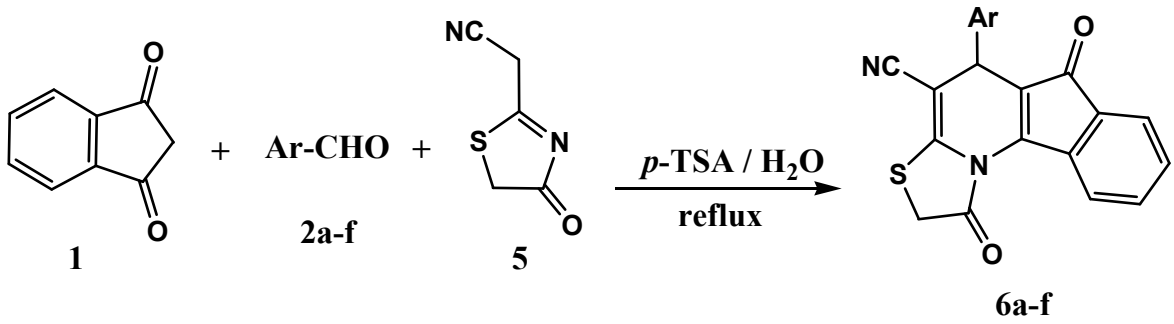


Scheme 1: Synthesis of novel indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**).

The elucidation of the structure of (**4**) using spectroscopic analysis is discussed with (**4b**) as representative example. The IR spectrum of (**4b**) showed absorption bands at ν_{max} = 2200 and 1730 cm^{-1} corresponding to CN and C=O groups respectively. The ^{1}H-NMR spectrum of (**4b**) exhibited one singlet signal at δ= 4.65 ppm for the methine proton and multiplet signals at δ= 7.40, 7.68 and 8.10 ppm for the aromatic protons of phenyl, indene and benzothiazole ring respectively. The ^{13}C-NMR spectrum of (**4b**) revealed the methine carbon (*CH*-7) at δ= 106.6, CN carbon at δ= 124.4 and carbonyl carbon at δ= 189.4 ppm. The mass spectrum shows the expected molecular ion peak.

The results were good in terms of yields and product purity in the presence of *p*-TSA, while without *p*-TSA the yields of products were trace even after very long reaction time (48 h).

To further expand the scope of the present method, we investigated one-pot, three-component reaction of 2-cyanomethyl-4-thiazolinone (**5**) instead of (benzothiazol-2-yl)acetonitrile (**3**) to obtain 10-aryl-11-cyanoindeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-diones (**6a-f**) in excellent yield (88-95%) (**Scheme 2**, **Table 1**).



Scheme 2: Synthesis of indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**).

The structure of the prepared compounds (**6a-f**) was confirmed from their elemental and spectral date. The IR spectrum of **6d** "as representative example" showed absorption bands at ν_{max} = 2200 and 1710, 1700 cm^{-1} corresponding to CN and two C=O groups respectively. The ^{1}H-NMR spectrum of (**6d**) exhibited two singlet signals at δ= 4.05 ppm for the CH_2 protons, 5.00 ppm for methine proton, multiplet signals at δ= 7.90 and 8.30 ppm for the aromatic protons of phenyl and

indene ring respectively, and broad signal at δ= 12.35 ppm for the toutomeric OH group. The ^{13}C-NMR spectrum of (**6d**) showed the following signals at δ= 35.2, 83.4 and 120.0 ppm for methylene (*CH*$_2$-2), methine (*CH*-10) and CN carbon respectively, in addition to, two signals at δ= 177.1, 189.6 ppm for two carbonyl carbons *C*-3, *C*-9 respectively. Also, the mass spectrum of (**6d**) shows the molecular ion peak at *m/z* = 404.86 corresponding (M^+ + 2) that established the molecular formula $C_{21}H_{11}N_3O_4S$.

The exact mechanism for the formation of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**) has not established, however, a reasonable possibility is shown in (**Scheme 3**).

Table 1: Synthesis of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**)

Product	***Ar***	***Time (h)***	***Yield (%)***[a]
4a	C_6H_5	7	75
4b	4-ClC_6H_4	4	89
4c	4-BrC_6H_4	4	83
4d	4-$O_2NC_6H_4$	3	81
4e	3-Pyridyl	6	79
4f	2-Thionyl	5	82
6a	C_6H_5	5	88
6b	4-ClC_6H_4	3	95
6c	4-BrC_6H_4	3	90
6d	4-$O_2NC_6H_4$	3	93
6e	3-Pyridyl	5	90
6f	2-Thionyl	4	91

[a] Isolated yield.

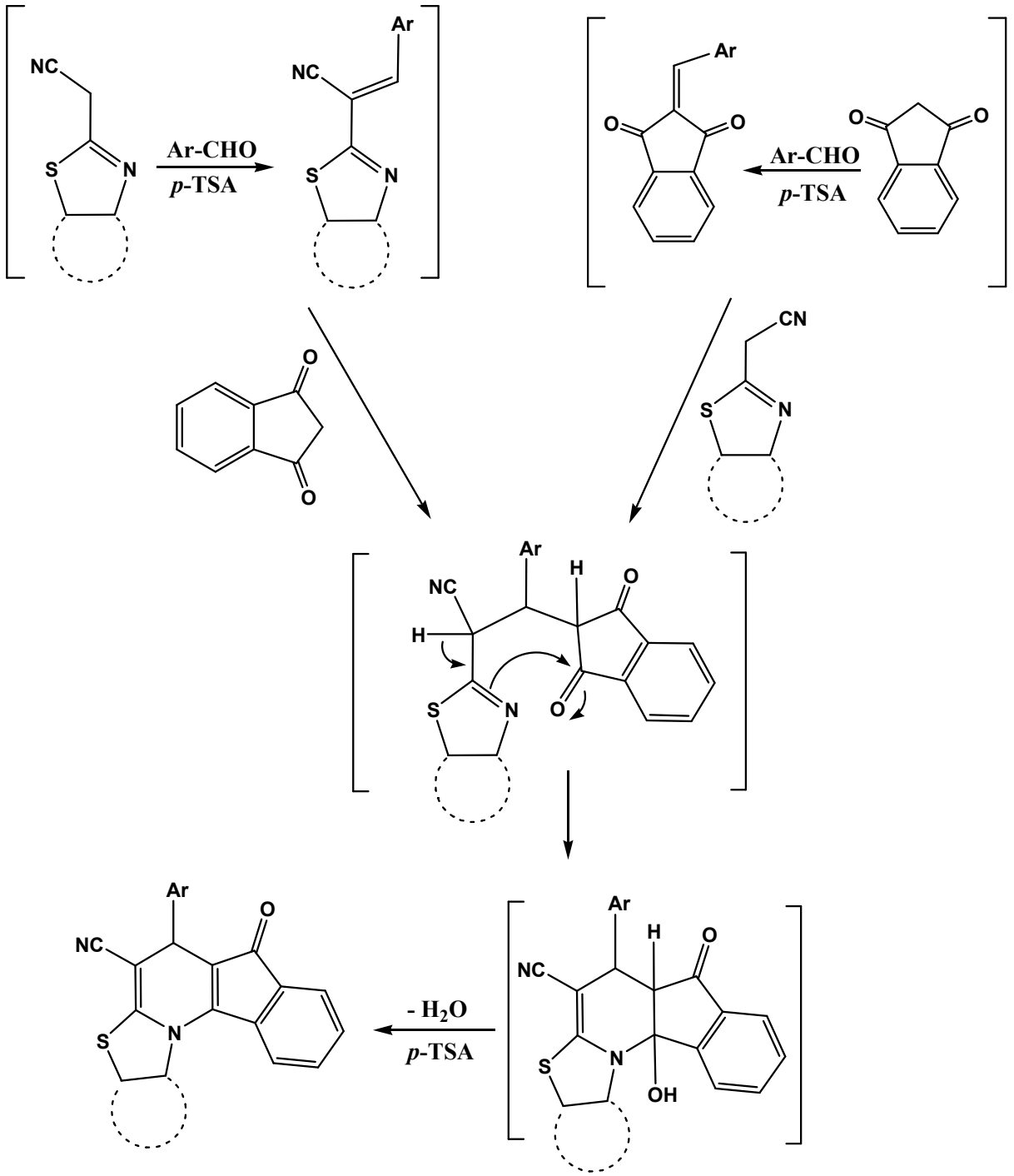
To the best of our knowledge, this new procedure provides the first example of an efficient and one-pot method for the synthesis of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines this method, based on three-component *p*-TSA-catalyzed reaction in water, is the most simple, convenient, clean and easy applicable.

Experimental Section

General: The time required for completion of each reaction was monitored by TLC. All melting points are uncorrected and were measured on a Gallenkamp apparatus. The IR spectra were recorded on a Shimadzu 470 IR spectrometer (KBr) ν_{max} / cm^{-1}. The ^{1}H, ^{13}C-NMR spectra were measured on Varian EM-200 (^{1}H: 200 MHz, ^{13}C: 50 MHz) spectrometer with TMS as internal standard and DMSO-d_6 as solvent. Mass spectra were determined on a JEOL JMS-600 spectrometer. Elemental analyses (C, H, N, and S) were performed on an elemental analysis system GmbH VarioEL $V_{2.3}$; the results were found to be in good agreement with the calculated values.

Synthesis of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (4a-f) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (6a-f).

General Procedure: A mixture of indan-1,3-dione **1** (0.146 g, 1 mmol), appropriate aromatic aldehyde **2a-f** (1 mmol), (benzothiazol-2-yl)acetonitrile **3** (0.174 g, 1 mmol) or 2-cyanomethyl-4-thiazolinone **5** (0.140 g, 1 mmol) in refluxing water (10 mL) containing catalytic amount of *p*-TSA (0.1 g) was stirred for the appropriate time (**Table 1**). Upon completion, monitored by TLC, the reaction mixture was allowed to cool to room temperature. The solid product was filtered off and washed with cold water (10 mL), ethanol (10 mL) and recrystallized for proper solvent to afford the pure product (**4a-f**) or (**6a-f**).



Scheme 3: The possible mechanism for the formation of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles (**4a-f**) and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines (**6a-f**).

6-Cyano-7-phenyl-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4a).
Recrystallize from dioxane as dense yellow crystals, m.p. 125-127 °C, yield 0.29 g (75%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1715 (C=O); ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.10 (s, 1H, CH), 7.45 (m, 4H, Ph-H), 8.00 (m, 4H, Ar-H-*indene*), 8.30 (m, 4H, Ar-H-*benzothiazole*). FAB MS: *m/z* (%)

390.00 (M^+, 1). Anal. Calcd. for $C_{25}H_{14}N_2OS$ (390.46): C, 76.90; H, 3.61; N, 7.17; S, 8.21. Found: C, 76.62; H, 3.40; N, 7.00; S, 8.00.

6-Cyano-7-(4-chlorophenyl)-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4b).
Recrystallize from dioxane as dense yellow crystals, m.p. 131-133 °C, yield 0.37 g (89%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1730 (C=O); ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.65 (s, 1H, CH), 7.40 (m, 4H, Ph-H), 7.68 (m, 4H, Ar-H-*indene*), 8.10 (m, 4H, Ar-H-*benzothiazole*). ^{13}C-NMR (DMSO-d_6): δ (ppm) 106.6 (CH), 115.9 (2C arom.), 124.4 (CN), 125.5, 125.9, 126.9, 127.4, 130.4, 131.8 (2CH arom.), 131.9 (2CH arom.), 132.6, 132.9, 134.2, 135.6, 136.2, 136.7 (2C arom.), 143.8, 146.1, 152.7, 162.3, 189.4 (CO). FAB MS: *m/z* (%) 424.50 (M^+, 2). Anal. Calcd. for $C_{25}H_{13}ClN_2OS$ (424.90): C, 70.67; H, 3.08; Cl, 8.34; N, 6.59; S, 7.55. Found: C, 70.50; H, 2.89; Cl, 8.15; N, 6.40; S, 7.36.

6-Cyano-7-(4-bromophenyl)-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4c).
Recrystallize from dioxane as yellow crystals, m.p. 138-140 °C, yield 0.38 g (83 %). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1720 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.20 (s, 1H, CH), 7.66 (m, 4H, Ph-H), 8.20 (m, 4H, Ar-H-*indene*), 8.50 (m, 4H, Ar-H-*benzothiazole*). ^{13}C-NMR (DMSO-d_6): δ (ppm) 106.1 (CH), 115.8 (2C arom.), 122.4 (CN), 123.1, 125.9, 126.3, 127.1, 129.7, 131.4 (2CH arom.), 131.8 (2CH arom.), 132.2, 132.3, 134.3, 135.4, 135.9, 136.0, 143.8, 146.8, 152.7, 162.8, 189.1 (CO). FAB MS: *m/z* (%) 470.00 (M^+, 5). Anal. Calcd. for $C_{26}H_{21}BrN_4O_3$ (469.35): C, 63.97; H, 2.79; Br, 17.02; N, 5.97; S, 6.83. Found: C, 63.81; H, 2.55; Br, 16.81; N, 5.70; S, 6.58.

6-Cyano-7-(4-nitrophenyl)-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4d).
Recrystallize from dioxane as pale brown crystals, m.p. 151-153 °C, yield 0.35 g (81%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1720 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.10 (s, 1H, CH), 7.48 (m, 4H, Ph-H), 8.05 (m, 4H, Ar-H-*indene*), 8.25 (m, 4H, Ar-H-*benzothiazole*). Anal. Calcd. for $C_{25}H_{13}N_3O_3S$ (435.45): C, 68.96; H, 3.01; N, 9.65; S, 7.36. Found: C, 68.81; H, 2.85; N, 9.45; S, 7.09.

6-Cyano-7-(3-pyridyl)-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4e).
Recrystallize from dioxane as yellow powder, m.p. 134-136 °C, yield 0.31 g (79%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1730 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.22 (s, 1H, CH), 7.71 (m, 4H, Ph-H), 8.28 (m, 4H, Ar-H-*indene*), 8.46 (m, 4H, Ar-H-*benzothiazole*). FAB MS: *m/z* (%) 392.01 (M^+, 1). Anal. Calcd. for $C_{24}H_{13}N_3OS$ (391.44): C, 73.64; H, 3.35; N, 10.73; S, 8.19. Found: C, 73.60; H, 3.10; N, 10.00; S, 7.92.

6-Cyano-7-(2-thionyl)-indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazole-8-one (4f).
Recrystallize from dioxane as yellow crystals, m.p. 143-145 °C, yield 0.32 g (82%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1730 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.15 (s, 1H, CH), 7.60 (m, 4H, Ph-H), 7.80 (m, 4H, Ar-H-*indene*), 8.25 (m, 4H, Ar-H-*benzothiazole*). FAB MS: *m/z* (%) 398.60 (M^+ + 2, 2). Anal. Calcd. for $C_{23}H_{12}N_2OS_2$ (396.48): C, 69.67; H, 3.05; N, 7.07; S, 16.17. Found: C, 69.55; H, 2.91; N, 6.86; S, 15.88.

11-Cyano-10-phenyl-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6a).
Recrystallize from ethanol as pale yellow crystals, mp 180-181 °C, yield 0.31 g (88%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1705 (C=O), 1700 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 3.95 (s, 2H, CH_2),

4.18 (s, 1H, CH), 7.30 (m, 4H, Ph-H), 8.10 (m, 4H, Ar-H-*indene*), 12.20 (s, 1H, *toutomeric* OH). Anal. Calcd. for $C_{21}H_{12}N_2O_2S$ (356.40): C, 70.77; H, 3.39; N, 7.86; S, 9.00. Found: C, 70.53; H, 3.10; N, 7.66; S, 8.80.

11-Cyano-10-(4-chlorophenyl)-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6b).
Recrystallize from ethanol as pale yellow crystals, m.p. 195-197 °C, yield 0.37 g (95%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1710 (C=O), 1700 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.00 (s, 2H, CH_2), 4.86 (s, 1H, CH), 7.39 (m, 4H, Ph-H), 8.15 (m, 4H, Ar-H-*indene*), 12.30 (s, 1H, *toutomeric* OH). ^{13}C-NMR (DMSO-d_6): δ (ppm) 33.7 (CH_2), 80.3 (CH), 81.3 (C), 115.9 (C), 117.7 (CN), 128.2, 128.6, 128.8 (2CH arom.), 129.2, 131.1 (2CH arom.), 131.6, 135.4, 135.9, 137.8, 141.9, 143.7, 155.8, 174.7 (CO),188.5 (CO). FAB MS: *m/z* (%) 390.16 (M^+, 2). Anal. Calcd. for $C_{21}H_{11}ClN_2O_2S$ (390.84): C, 64.53; H, 2.84; Cl, 9.07; N, 7.17; S, 8.20. Found: C, 63.29; H, 2.74; Cl, 8.90; N, 7.00; S, 8.00.

11-Cyano-10-(4-bromophenyl)-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6c).
Recrystallize from ethanol as pale yellow crystals, m.p. 163-165 °C, yield 0.39 g (90%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1705 (C=O), 1700 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.00 (s, 2H, CH_2), 4.19 (s, 1H, CH), 7.45 (m, 4H, Ph-H), 8.00 (m, 4H, Ar-H-*indene*), 12.10 (s, 1H, *toutomeric* OH). FAB MS: *m/z* (%) 437.50 (M^+ + 2, 2). Anal. Calcd. for $C_{21}H_{11}BrN_2O_2S$ (435.29): C, 57.94; H, 2.55; Br, 18.36; N, 6.44; S, 7.37. Found: C, 57.68; H, 2.15; Br, 18.15; N, 6.40; S, 7.07.

11-Cyano-10-(4-nitrophenyl)-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6d).
Recrystallize from ethanol as pale yellow crystals, m.p. 191-193 °C, yield 0.37 g (93%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1710 (C=O), 1700 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.05 (s, 2H, CH_2), 5.00 (s, 1H, CH), 7.90 (m, 4H, Ph-H), 8.30 (m, 4H, Ar-H-*indene*), 12.35 (s, 1H, *toutomeric* OH). ^{13}C-NMR (DMSO-d_6): δ (ppm) 35.2 (CH_2), 83.4 (CH), 84.1 (C), 116.3 (C), 120.0 (CN), 128.6, 129.2, 129.8 (2CH arom.), 129.8, 132.6 (2CH arom.), 133.1, 135.9, 136.9, 138.2, 143.9, 144.9, 157.7, 177.1 (CO),189.6 (CO). FAB MS: *m/z* (%) 404.86 (M^+ + 2, 1). Anal. Calcd. for $C_{21}H_{11}N_3O_4S$ (401.39): C, 62.84; H, 2.76; N, 10.47; S, 7.99. Found: C, 62.69; H, 2.56; N, 10.38; S, 7.85.

11-Cyano-10-(3-pyridyl)-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6e).
Recrystallize from ethanol as yellow crystals, m.p. 174-176 °C, yield 0.32 g (90%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1715 (C=O), 1705 (CO), 1680 (C=N). ^{1}H-NMR (DMSO-d_6): δ (ppm) 4.10 (s, 2H, CH_2), 4.98 (s, 1H, CH), 7.75 (m, 4H, Ph-H), 8.61 (m, 4H, Ar-H-*indene*). FAB MS: *m/z* (%) 353.10 (M^+, 2). Anal. Calcd. for $C_{20}H_{11}N_3O_2S$ (357.39): C, 67.21; H, 3.10; N, 11.76; S, 8.97. Found: C, 66.97; H, 2.96; N, 11.50; S, 8.80.

11-Cyano-10-(2-thionyl)-indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridine-3,9-dione (6f).
Recrystallize from ethanol as yellow crystals, m.p. 150-152 °C, yield 0.33 g (91%). IR (KBr): ν_{max}/cm^{-1} 2200 (CN), 1710 (C=O), 1700 (CO). ^{1}H-NMR (DMSO-d_6): δ (ppm) 3.96 (s, 2H, CH_2), 5.00 (s, 1H, CH), 7.30 (m, 4H, Ph-H), 8.10 (m, 4H, Ar-H-*indene*). FAB MS: *m/z* (%) 364.30 (M^+ + 2, 3). Anal. Calcd. for $C_{19}H_{10}N_2O_2S_2$ (362.42): C, 62.97; H, 2.78; N, 7.73; S, 17.69. Found: C, 62.75; H, 2.61; N, 7.75; S, 17.60.

Conclusion

In conclusion, I have described a facile, one-pot, three-component and green method for the synthesis of indeno[2`,3`:5,6]pyrido[2,1-*b*]benzothiazoles and indeno[2`,3`-*e*]thiazolo[3,2-*a*]pyridines in water, prominent among the advantages of this new method are novelty, operational simplicity, good yields and easy work-up procedures employed.

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