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Novel 1,3-Dithiolan-2-ones Using Sodium a-Cyanoketene Dithiolates

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Novel 1,3-Dithiolan-2-ones Using Sodium α-Cyanoketene Dithiolates

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Abstract: A novel and efficient method for the synthesis of 1,3-dithiolan-2-ones using sodium α -cyanoketene dithiolates and monochloroacetic acid as starting components is described.

Keywords: α -Cyanoketene dithioacetals, 1,3-dithiolan-2-ones, heterocycles, monochloroacetic acid, sodium α -cyanoketene dithiolates

Recently, we reported an improved method for the preparation of novel α cyanoketene *S*,*S*-acetals **2** and their conversions to various substituted methylthio derivatives of pyrrole, pyrazole, pyridine, and pyrimidine.^[1] In an extension of this work, we now report a novel synthesis of functionalized 1,3-dithiolanes using sodium α -cyanoketene dithiolate **3** as versatile precursors (Scheme 1). Thus, it has been found that substituted acetanilide derivatives **la**–**d** reacted with carbon disulphide in the presence of sodium ethoxide to give the corresponding sodium α -cyanoketene dithiolates **3**. The latter were alkylated with chloroacetic acid to give the novel monoalkylated products **7**. The structures of **7** were established on the basis of their elemental analysis and spectral data (IR, MS, ¹H NMR). On reflux in ethanolic sodium ethoxide compounds **7** undergo cyclization to give the

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corresponding 1,3-dithiolane derivatives **8**. The structure of compounds **8** was established on the basis of their elemental analysis and spectral data. Compounds **8** can also be prepared by alkylation of the dithiolate **3** with ethyl bromoacetate followed by cyclization of the resulting acyclic product

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6. Compounds 8 bear an active methylene function adjacent to a carbonyl group and are expected to react with unsaturated nitriles to give condensed dithiolane derivatives. Thus, it has been found that dithiolanes 8 reacted with [bis(methylthio)methylene]malononitrile 9 in refluxing ethanol containing catalytic amounts of piperidine to give *N*-aryl-4-methylsulfanyl-2-pyridones 10a-d and not the expected pyranodithiolane derivatives 13 (Scheme 2). The structures of 10 were established on the basis of their elemental analysis and spectral data (MS, IR, ¹H NMR). Moreover, the products 10 obtained were shown to be identical to those obtained from our



Scheme 2.

previously reported reaction of 1 with 9 by their melting points and spectral data.^[2] The formation of 10 from 1 and 9 is assumed to proceed via the intermediacy of 12. The unexpected course of the reaction between dithiolanes 8 and the ketene dithioacetal 9 prompted us to investigate this reaction for the dithiolanes 8 and the arylmethyenemalononitriles 14 under the same conditions (Scheme 3). Similarly, the *N*-aryl-2-pyridone products 15 were obtained, identical to those obtained from the reaction of cyanoacetanilides 1 with arylmethylenemalononitriles 14 (melting points, spectral data) (Table 1).^[3-6] The mechanism of the reaction of 1 and 14 is assumed to proceed through the formation of the initial adduct 17, which leads to the product 15 as produced by the reaction of 8 with 14. Although an investigation of the reaction mechanism for the formation of 10 and 15 through route (A) was not undertaken, the reaction pathway is considered to proceed via



Scheme 3.

Ar	Ar ¹
C ₆ H ₅	$4-CH_3-C_6H_4$
C_6H_5	$4-OCH_3-C_6H_4$
C_6H_5	$4-Cl-C_6H_4$
$4-CH_3-C_6H_4$	C_6H_5
$4-CH_3-C_6H_4$	$4-CH_3-C_6H_4$
$4-CH_3-C_6H_4$	$4-OCH_3-C_6H_4$
$4-CH_3-C_6H_4$	$4-Cl-C_6H_4$
4-OCH ₃ -C ₆ H ₄	C_6H_5
4-OCH ₃ -C ₆ H ₄	$4-CH_3-C_6H_4$
$4-OCH_3-C_6H_4$	$4-OCH_3-C_6H_4$
$4-OCH_3-C_6H_4$	$4-Cl-C_6H_4$
$4-Cl-C_6H_4$	C_6H_5
$4-Cl-C_6H_4$	$4-CH_3-C_6H_4$
$4-Cl-C_6H_4$	4-OCH ₃ -C ₆ H ₄
$4-Cl-C_6H_4$	$4-Cl-C_6H_4$
	$\begin{tabular}{lllllllllllllllllllllllllllllllllll$

cleavage of the exocyclic double bond under the reaction conditions in the dithiolanes 8 and cyanoacetanilide elimination. The latter reacts with 9 or 14 to yield products 10 or 15, respectively. Similar reactions were previously reportect.^[7-10]

In summary, we have achieved a regiospecifc synthesis of interesting novel 1,3-dithiolane derivatives by the reaction of sodium α -cyanoketene dithiolates with chloroacetic acid followed by cyclization. The compounds obtained seem promising for further chemical transformations.

EXPERIMENTAL

All melting points were determined on an electrothermal melting-point apparatus and are uncorrected. IR (cm⁻¹) spectra were recorded on KBr disk on a FT IR-8201 PC Schimadzu spectrophotometer. ¹H NMR spectra were recorded in CDCl₃ or (CD₃)₂SO on a Gemini 200-MHz spectrophotometer using TMS as internal reference, and chemical shifts are expresses as δ ppm unit. Mass spectroscopy was recorded on a GC-MS QP1000 EX Shimadzu. Elemental analyses were performed at the microanalytical center at Cairo University.

Ethyl {[3-(Arylamino)-2-cyano-l-mercapto-3-oxoprop-1-enyl]thio} acetate (6a-d), General Procedure

Ethyl bromoacetate (1.67 gm, 0.01 mol) was added in portions at room temperature to a solution of compounds 3a-d (0.01 mol) in ethanol (20 mL). The reaction mixture was left for 2 h under stirring, then poured over iced water,

and neutralized with dilute hydrochloric acid. The precipitated product was filtered off and recrystallized from absolute ethanol (20 mL).

6a: Yellow, mp 203 °C; yield 90%. ν_{max}/cm^{-1} (KBr), 3271 (NH); 2206 (CN), 1735 (CO), 1670 (CO) cm⁻¹; ¹H NMR (DMSO) δ 1.97 (t, 3H, CH₃), 3.74 (s, 2H, CH₂), 4.87 (q, 2H,CH₂), 7.71–7.96 (m, 5H, C₆H₅), 9.21 (s, 1H, NH), and 10.64 (s, 1H, SH). Found: C, 52.35; H, 4.61; N, 8.43%. Calcd. for C₁₄H₁₄N₂O₃S₂: C, 52.17; H, 4.35; N, 8.70%.

6b: Yellow, mp 220 °C, yield 80%. ν_{max}/cm^{-1} (KBr), 3282 (NH), 2202 (CN), 1735 (CO), 1635 (CO) cm⁻¹; ¹H NMR (DMSO) δ 1.59 (t, 3H, CH₃), 2.66 (s, 3H, CH₃), 4.37 (s, 2H, CH₂), 4.54 (q, 2H,CH₂), 7.22–7.62 (m, 4H, C₆H₄), 8.12 (s, IH, NH); m/z 336. Found: C, 53.82; H, 4.82; N, 8.51%. Calcd. for C₁₅H₁₆N₂O₃S₂: C, 53.57; H, 4.76; N, 8.33%.

6c: Yellow, mp 235 °C, yield 85%. ν_{max}/cm^{-1} (KBr), 3275 (NH); 2200 (CN), 1700 (CO), 1660 (CO) cm⁻¹; ¹H NMR (DMSO) δ 1.36 (t, 3H, CH₃), 2.82 (s, 3H, OCH₃), 3.16 (s, 2H, CH₂), 4.33 (q, 2H, CH₂), 6.90–7.46 (m, 4H, C₆H₄), 7.99 (s, 1H, NH), 12.00 (s, 1H, SH). Found: C, 51.41; H, 4.61; N, 7.63%. Calcd. for C₁₅H₁₆N₂O₄S₂: C, 51.14; H, 4.55; N, 7.95%.

6d: Yellow, mp 233 °C, yield 89%. ν_{max}/cm^{-1} (KBr), 3420 (NH); 2203 (CN), 1695 (CO), 1670 (CO) cm⁻¹; ¹H NMR (DMSO) δ 1.23 (t, 3H, CH₃), 3.63 (s, 2H, CH₂), 4.17 (q, 2H, CH₂), 7.38–7.63 (m, 4H, C₆H₄), 8.85 (s, 1H, NH), 10.41 (s, 1H, SH); m/z 358. Found: C, 47.41; H, 3.43; N, 7.58%. Calcd. for C₁₄H₁₃N₂O₃S₂Cl: C, 47.12; H, 3.65; N, 7.85%.

{[3-(Arylamino)-2-cyano-1-mercapto-3-oxoprop-1-enyl]thio}acetic Acids 7a-d, General Procedure

 α -Chloroacetic acid (0.94 gm, 0.01 mol) was added in portions to a solution of compounds **3a-d** (0.01 mol) in ethanol (20 mL) at room temperature. The reaction mixture was left for 30 min under stirring, then poured over an ice-water mixture, and neutralized with dilute hydrochloric acid. The precipitated product was filtered off and recrystallized from ethanol.

7a: Yellow, mp 190 °C, yield 92%. $\nu_{\rm m}$.cm⁻¹(KBr), 3551, 3417 (OH, NH); 2202 (CN), 1690 (CO), 1675 (CO) cm⁻¹. Found: C, 48.63; H, 3.62; N, 9.31% Calcd. for C₁₂H₁₀N₂O₃S₂: C, 48.98; H, 3.40; N, 9.52%.

7b: Yellow, mp 245 °C, yield 84%. ν_{max}/cm^{-1} (KBr), 3460, 3340 (OH, NH); 2200 (CN), 1697 (CO), 1670 (CO) cm^{-1} ; ¹H NMR (DMSO) δ 2.29 (s, 3H, CH₃), 4.33 (s, 2H, CH₂), 6.20–7.65 (m, 4H, C₆H₄), 9.50 (s, 1H, NH), 9.90 (s, 1H, SH), 12.31 (s, 1H, OH); m/z 307. Found: C, 50.42; H, 3.72; N, 9.21%. Calcd. for C₁₃H₁₂N₂O₃S₂: C, 50.65; H, 3.90; N, 9.09%.

7c: Yellow, mp 205 °C, yield 81%. ν_{max}/cm^{-1} (KBr), 3400, 3330 (OH, NH); 2211 (CN), 1695 (CO), 1655 (CO) cm⁻¹; ¹H NMR (DMSO) δ 3.97 (s, 3H,

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OCH₃), 4.62 (s, 2H, CH₂), 7.10–7.64 (m, 4H, C₆H₄), 9.88 (s, 1H, NH), 10.05 (s, 1H, SH), 10.36 (s, 1H, OH); m/z 326. Found: C, 48.43; H, 3.52; N, 8.42%. Calcd. for $C_{13}H_{12}N_2O_4S_2$: C, 48.15; H, 3.70; N, 8.64%.

7d: Yellow, mp 220 °C, yield 80%. ν_{max}/cm^{-1} (KBr), 3460–2500 (OH, NH); 2200 (CN), 1697 (CO), 1670 (CO) cm⁻¹; ¹H NMR (DMSO) δ 3.96 (s, 2H, CH₂), 6.34–7.62 (m, 4H, C₆H₄), 10.29 (s, 1H, NH), 10.48 (s, 1H, SH), 12.99 (s, 1H, OH); m/z 331. Found: C, 43.61; H, 2.52; N, 8.33%. Calcd. for C₁₂H₉N₂O₃S₂Cl: C, 43.84; H, 2.74; N, 8.52%.

N-Aryl-2-cyano-2-(4-oxo-1,3-dithiolan-2-ylidene)-acetamides (8a–d), General Procedure

Compounds 6a-d or 7a-d (0.01 mol) were heated in sodium ethoxide (0.046 g 0.01 mol) and ethanol (30 mL) for 30 min and then cooled. The mixture was poured over an ice-water mixture and neutralized with dilute hydrochloric acid, and the precipitated product was filtered off and recrystal-lized from the appropriate solvent.

8a: Yellow, from ethanol, mp 190 °C, yield 75%. ν_{max}/cm^{-1} (KBr) 3290 (NH), 2205 (CN), 1679 (CO), 1658 (CO) cm⁻¹; ¹H NMR (DMSO) δ 4.40 (s, 2H, CH₂), 7.26–7.55 (m, 5H, C₆H₅), 12.00 (s, 1H, NH); m/z 276. Found: C, 52.32; H, 3.01; N, 10.32%. Calcd. for C₁₂H₈N₂O₂S₂: C, 52.17; H, 2.89; N, 10.14%.

8b: Yellow, from ethanol, mp 165 °C, yield 79%. ν_{max}/cm^{-1} (KBr), 3280 (NH), 2206 (CN), and 1742 (CO), 1636 (CO) cm⁻¹; ¹H NMR (DMSO) δ 2.23 (s, 3H, CH₃), 3.67 (s, 2H, CH₂), 7.12–7.55 (m, 4H, C₆H₄), 12.44 (s, 1H, NH); m/z 290. Found: C, 53.63; H, 3.61; N, 9.81%. Calcd. for C₁₃H₁₀N₂O₃S₂: C, 53.79; H, 3.45; N, 9.66%.

8c: Yellow, from ethanol, mp 185 °C yield 85%. ν_{max}/cm^{-1} (KBr), 3327 (NH), 2208 (CN), and 1685 (CO), 1660 (CO) cm⁻¹. Found: C, 51.01; H, 3.42; N, 9.32%. Calcd. for $C_{13}H_{10}N_2O_3S_2$: C, 50.98; H, 3.26; N, 9.15%.

8d: Yellow, from EtOH, mp 250 °C, yield 80%. ν_{max}/cm^{-1} (KBr), 3316 (NH), 2203 (CN), 1680, 1656 (CO) cm⁻¹. Found: C, 46.62; H, 2.41; N, 9.31%. Calcd. for C₁₂H₇N₂O₂S₂Cl: C, 46.45; H, 2.25; N, 9.03%.

6-Amino-1-aryl-4-(methylthio)-2-oxo-1,2-dihydropyridine-3,5dicarbonitriles (10a-d)

Method (a): General Procedure

A mixture of 8a-d (0.01 mol) and bis(methylthiomethylene)malononitrile 9 (1.70 g, 0.01 mol) was boiled under reflux in ethanol (30 mL) containing a

catalytic amount of piperidine for 30 min and then cooled. The precipitated product was filtered off and recrystallized from ethanol.

Method (b): General Procedure

A mixture of bis(methylthiomethylene)malononitrile 9 (1.70 g, 0.01 mol) and *N*-arylcyanoacetamide derivatives 1 was heated in ethanol (40 mL) in the presence of catalytic amounts of piperidine for 1 h and then cooled. The precipitated product was filtered off and recrystallized from ethanol.

6-Amino-1,4-diaryl-2-oxo-1,2-dihydropyridin-3,5-dicarbonitriles (15a-0)

Method (a): General Procedure

A mixture of compounds **8** (0.01 mol) and arylmethylenemalononitriles **14** (0.01 mol) was refluxed in ethanol (30 mL) in the presence of catalytic amounts of piperidine for 30 min and then cooled. The precipitated product was filtered off and recrystallized from ethanol.

Method (b): General Procedure

A mixture of arylmethylenemalononitriles **14** (0.01 mol) and *N*-aryl-cyanoacetamide derivatives **1** (0.01 mol) was refluxed in ethanol (30 mL) containing catalytic amounts of piperidine (3 drops) for 20 min and then cooled. The formed product was filtered off and recrystallized from ethanol.

15a: Buff, mp > 300 °C, yield 70%. ν_{max}/cm^{-1} (KBr), 3440, 3190 (NH₂), 2214 (2CN), and 1654 (CO) cm⁻¹; ¹H NMR (DMSO) δ 2.51 (s, 3H, CH₃), 7.25–7.58 (m, 9H, C₆H₄ and C₆H₅), 7.79 (s, 2H, NH₂); m/z 326. Found: C, 73.76; H, 3.99; N, 17.42%. Calcd. for C₂₀H₁₄N₄O: C, 73.62; H, 4.29; N, 17.18%.

15b: Buff, mp > 300 °C, yield 75%. ν_{max}/cm^{-1} (KBr), 3421, 3193 (NH₂), 2210 (2CN), and 1666 (CO) cm⁻¹. Found: C, 70.43; H, 4.11; N, 16.45%. Calcd. for C₂₀H₁₄N₄O₂: C, 70.18; H, 4.09; N, 16.37%.

15c: Yellow, mp > 300 °C, yield 70%. ν_{max}/cm^{-1} (KBr), 3440, 3197 (NH₂), 2218 (2CN), and 1635 (CO) cm⁻¹; ¹H NMR (DMSO) δ 7.45–7.70 (m, 9H, C₆H₄ and C₆H₅), 8.00 (s, 2H, NH₂); m/z 346. Found: C, 65.66; H, 3.36; N, 16.51% Calcd. for C₁₉H₁₁N₄OCl: C, 65.90; H, 3.18; N, 16.18%.

15d: Buff, mp > 300 °C, yield 75%. ν_{max}/cm^{-1} (KBr), 3445, 3190 (NH₂), 2213 (2CN), 1660 (CO) cm⁻¹. Found: C, 73.46; H, 3.99; N, 17.22%. Calcd. for C₂₀H₁₄N₄O: C, 73.62; H, 4.29; N, 17.18%.

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15e: Yellow, mp > 300 °C, yield 71%. ν_{max}/cm^{-1} (KBr), 3456, 3201 (NH₂), 2221 (2CN), and 1654 (CO) cm⁻¹; ¹H NMR (DMSO) δ 2.40 (s, 3H, CH₃), 2.42 (s, 3H, CH₃), 7.25–7.49 (m, 8H, 2C₆H₄), 7.81 (s, 2H, NH₂); m/z 340. Found: C, 74.35; H, 4.55; N, 16.63%. Calcd. for C₂₁H₁₆N₄O: C, 74.12; H, 4.71; N, 16.97%.

15f: Yellow, mp > 300 °C, yield 65%. ν_{max}/cm^{-1} (KBr), 3429, 3201 (NH₂), 2221 (2CN), 1635 (CO) cm⁻¹; Found: C, 70.53; H, 4.31; N, 15.64%. Calcd. for C₂₁H₁₆N₄O₂: C, 70.79; H, 4.49; N, 15.73%.

15g: Buff, mp > 300 °C, yield 63%. ν_{max}/cm^{-1} (KBr), 3435, 3197 (NH₂), 2210 (2CN), 1650 (CO) cm⁻¹. Found: C, 66.45; H, 3.72; N, 15,43%. Calcd. for C₂₀H₁₃N₄OCl: C, 66.66; H, 3.61; N, 15.55%.

15h: Yellow, mp > 300 °C, yield 75%. ν_{max}/cm^{-1} (KBr), 3445, 3201 (NH₂), 2216 (2CN), 1644 (CO) cm⁻¹. Found: C, 70.32; H, 4.22; N, 16.33%. Calcd. for C₂₀H₁₄N₄O₂: C, 70.18; H, 4.09; N, 16.37%:

15i: Buff, mp > 300 °C, yield 69%. ν_{max}/cm^{-1} (KBr), 3456, 3197 (NH₂), 2221 (2CN), and 1651 (CO) cm⁻¹; ¹H NMR (DMSO) δ 2.42 (s, 3H, CH₃), 3.87 (s, 3H, OCH₃) 7.12–7.55 (m, 8H, 2C₆H₄), 7.74 (s, 2H, NH₂); m/z 356. Found: C, 70.64; H, 4.43; N, 15.54%. Calcd. for C₂₁H₁₆N₄O₂: C, 70.79; H, 4.49; N, 15.73%.

15j: Yellow, mp > 300 °C, yield 75%. ν_{max}/cm^{-1} (KBr), 3398, 3190 (NH₂), 2210 (2CN), and 1658 (CO) cm⁻¹; ¹H NMR (DMSO) δ 3.80 (s, 3H, OCH₃), 3.85 (s, 3H, OCH₃), 7.12–7.55 (m, 8H, 2C₆H₄), 7.76 (s, 2H, NH₂). Found: C, 67.55; H, 4.31; N, 15.21%. Calcd. for C₂₁H₁₆N₄O₃: C, 67.74; H, 4.30; N, 15.05%.

15k: Yellow, mp > 300 °C, yield 72%. ν_{max}/cm^{-1} (KBr), 3430, 3195 (NH₂), 2214 (2CN), and 1652 (CO) cm⁻¹. Found: C, 63.77; H, 3.22; N, 15.11%. Calcd. for C₂₀H₁₃N₄O₂Cl: C, 63.83; H, 3.46; N, 14.89%.

15I: Buff, mp > 300 °C, yield 65%. ν_{max}/cm^{-1} (KBr), 3440, 3198 (NH₂), 2218 (2CN), and 1644 (CO) cm⁻¹. Found: C, 65.51; H, 3.26; N, 16.22%. Calcd. for C₁₉H₁₁N₄OCl: C, 65.90; H, 3.18; N, 16.18%.

15m: Buff, mp > 300 °C, yield 61%. ν_{max}/cm^{-1} (KBr), 3398, 3193 (NH₂), 2214 (2CN), and 1651 (CO) cm⁻¹. Found: C, 66.57; H, 3.55; N, 15.62%. Calcd. for C₂₀H₁₃N₄OCl: C, 66.66; H, 3.61; N, 15.55%.

15n: Buff, mp > 300 °C, yield 64%. ν_{max}/cm^{-1} (KBr), 3398, 3193 (NH₂), 2214 (2CN), 1655 (CO) cm⁻¹; ¹H NMR (DMSO) δ 3.87 (s, 3H, OCH₃) 7.13–7.66 (m, 8H, 2C₆H₄), 7.94 (s, 2H, NH₂). Found: C, 63.56; H, 3.32; N, 14.77%. Calcd. for C₂₀H₁₃N₄O₂Cl: C, 63.83; H, 3.46; N, 14.89%.

150: Yellow, mp > 300 °C, yield 76%. ν_{max}/cm^{-1} (KBr), 3398, 3194 (NH₂), 2214 (2CN), 1651 (CO) cm⁻¹. Found: C, 59.66; H, 2.54; N, 14.53%. Calcd. for C₂₀H₁₃N₄O₂Cl₂: C, 59.84; H, 2.62; N, 14.70%.

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