A Convenient Metal-Free Method for the Synthesis of Benzothiazolethiones from *o*-Haloanilines and Carbon Disulfide

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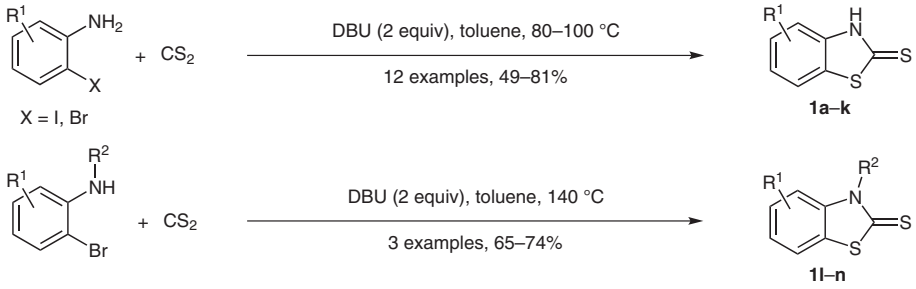
Abstract: A convenient method has been developed for the preparation of a variety of 1,3-benzothiazole-2(3*H*)-thiones. The reaction proceeds from an *o*-haloaniline derivative and carbon disulfide in the presence of 1,8-diazabicyclo[5.4.0]undec-7-ene at 80–140 °C to give the corresponding 1,3-benzothiazole-2(3*H*)-thione derivatives in good-to-excellent yields.

Key words: cyclizations, heterocycles, benzothiazoles, amines, carbon disulfide, tandem reaction



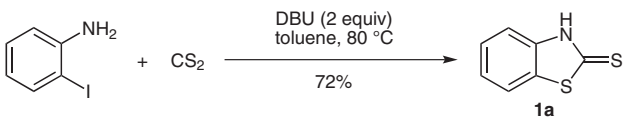
Scheme 1 Synthesis of various substituted 1,3-benzothiazole-2(3*H*)-thiones

Introduction

1,3-Benzothiazole-2(3*H*)-thiones[1] form the core structures of pharmaceutical agents[2] and advanced materials.[3] Classical methods for the preparation of 1,3-benzothiazole-2(3*H*)-thiones included the reactions of *N*,*N*′-diphenylthioureas with sulfur or the reactions of 2-aminobenzenethiols with carbon disulfide under high pressure.[4] Another way of preparing 1,3-benzothiazole-2(3*H*)-thiones is by means of a nucleophilic aromatic substitution (S_NAr) reaction of a potassium or sodium *O*-ethyldithiocarbonate with an *o*-haloaniline followed by cyclization.[5] Some of the reported methods for the preparation of 1,3-benzothiazole-2(3*H*)-thiones have the drawbacks of requiring harsh conditions, of having a limited substrate scope, of showing poor tolerance of substituents, or of requiring the use of transition-metal catalysts. Accordingly, the development of a simple and scalable route for constructing the 1,3-benzothiazole-2(3*H*)-thione skeleton is desirable. We have recently reported an efficient strategy for the preparation of 1,3-benzothiazole-2(3*H*)-thione derivatives from *o*-haloaniline derivatives and carbon disulfide through a tandem reaction in the presence of 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU).[6] This reaction tolerates a broad spectrum of substituents and occurs under mild and metal-free conditions. Here, we report our efforts to prepare various 1,3-benzothiazole-2(3*H*)-thiones on a 5-mmol scale by using this method (Scheme 1). The reaction can also be used to prepare N-substituted 1,3-benzothiazole-2(3*H*)-thiones.

Scope and Limitations

2-Iodoaniline and carbon disulfide are commercially available. The reaction of 2-iodoaniline (1 equiv) with carbon disulfide (2 equiv) in the presence of 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU; 2 equiv) as a base at 80 °C for 24 hours gave 1,3-benzothiazole-2(3*H*)-thione in 72% isolated yield, as shown in Scheme 2.



Scheme 2 Reaction of *o*-iodoaniline with carbon disulfide

2-Bromoaniline and its derivatives are cheaper and more readily available than the corresponding 2-iodoaniline derivatives. For these reasons, we examined a range of 2-bromoanilines as reactants for the synthesis of 1,3-benzothiazole-2(3*H*)-thiones (Table 1). Although a slightly

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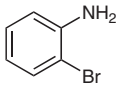
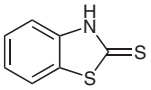
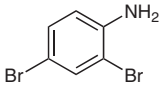
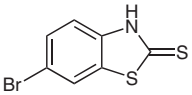
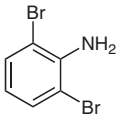
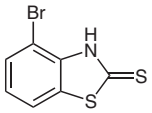
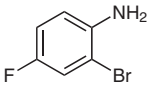
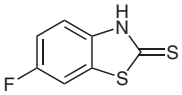
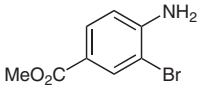
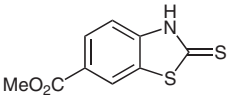
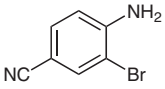
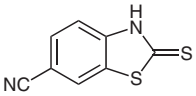
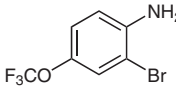
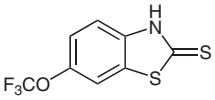
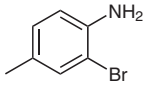
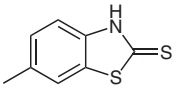
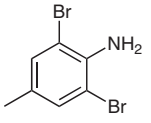
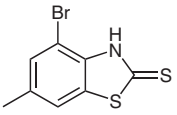
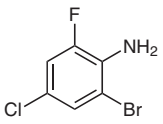
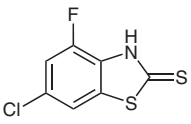
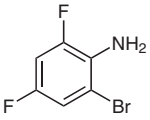
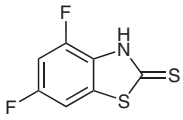
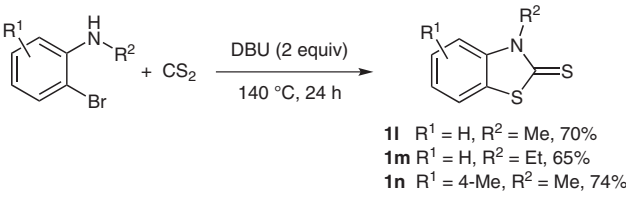
higher temperature (100 °C) was required to complete the reaction, 2-bromoanilines with either electron-donating or electron-withdrawing substituents on the ring gave good-to-excellent yields of the corresponding 1,3-benzothiazole-2(3*H*)-thiones. The range of *s*ubstituents tolerated ranged from weakly electron-withdrawing groups (such as bromo) to strongly electron-withdrawing groups (such as methoxycarbonyl, trifluoromethoxy, or cyano), as well as electron-donating substituents (methyl) in the *para*-position. In each case, the corresponding substituted 1,3-benzothiazole-2(3*H*)-thione was obtained in a satisfactory yield (entries 2–8). Substrate with two substituents located *para* and *ortho* to the amino group also gave high yields of the corresponding 1,3-benzothiazole-2(3*H*)-thiones (entries 9–11). In the case of 2-chloroaniline, no reaction occurred with carbon disulfide, even when the temperature was increased to 120 °C.

Table 1 Reaction of 2-Iodoanilines with Carbon Disulfide

Entry	Reactant	Product	Yield[a] (%)
1		**1a** 	77
2		**1b** 	49
3		**1c** 	59
4		**1d** 	81
5		**1e** 	76
6		**1f** 	57
7		**1g** 	75
8		**1h** 	71
9		**1i** 	58
10		**1j** 	81
11		**1k** 	80

[a] Isolated yield.

Several N-substituted 1,3-benzothiazole-2(3*H*)-thiones have useful medicinal properties.[7] However, preparations of N-substituted 1,3-benzothiazole-2(3*H*)-thiones are limited by the need for harsh conditions[8] or because they give low yields.[9] When we applied our present strategy to the synthesis of 3-methylbenzothiazole-2(3*H*)-thione (**1l**), we obtained the product in 70% yield by performing the reaction at 140 °C (Scheme 3); 3-ethylbenzothiazole-2(3*H*)-thione (**1m**) was similarly obtained in 65% yield. When 2-bromo-*N*,4-dimethylaniline was used as the reactant, the dimethylated product **1n** was obtained in 74% yield.



Scheme 3 Reactions of N-substituted bromoanilines with carbon disulfide

In summary, we have demonstrated a simple, practical, and highly efficient base-promoted method for the synthesis of 1,3-benzothiazole-2(3*H*)-thione and its derivatives. The protocol uses readily available 2-iodo- or 2-bromoanilines and carbon disulfide as the starting materials, requires mild conditions, and gives the corresponding 1,3-benzothiazole-2(3*H*)-thiones in good-to-excellent yields. The method eliminates the need for any metal catalysts and the resulting products are of potential interest in both academic and pharmaceutical research.

All the reactions were carried out under N_2 in dried screw-cap tubes fitted with a Teflon-lined septa. Unless otherwise indicated, all materials were obtained from commercial sources and used as received. Toluene was freshly distilled. Column chromatography was performed on silica gel (particle size 10–40 μm; Ocean Chemical Factory, Qingdao, China). Common solvents for chromatography, such as petroleum ether (PE) and EtOAc, were purchased from commercial suppliers and used without further purification. ^{1}H NMR and ^{13}C NMR spectra were recorded on a JEOL AL-300 MHz, JEOL AL-600 MHz, or Bruker 400 MHz NMR spectrometers at ambient temperature with DMSO-d_6 as the solvent. The melting points were measured on X-4 digital melting point apparatus and are uncorrected. Mass spectra were recorded by using a Bruker Esquire ion-trap mass spectrometer in the positive-ion mode.

1,3-Benzothiazole-2(3*H*)-thione (1a); Typical Procedure

CAUTION: *Carbon disulfide is extremely flammable, and toxic by inhalation, skin absorption, and ingestion*.

A sealed tube (50 mL) was charged with 2-$BrC_6H_4NH_2$ (0.86 g, 5 mmol), DBU (1.5 mL, 10.0 mmol), and CS_2 (0.6 mL, 10 mmol). Toluene (8 mL) at r.t. was added under N_2 and, after 30 min, the tube was sealed and the mixture was stirred at 100 °C for 24 h. The mixture was then cooled to r.t. and H_2O (20 mL) was added. The

mixture was extracted with EtOAc (3 × 15 mL) and the extracts were dried (Na_2SO_4) and concentrated. The residue was purified by chromatography [silica gel, PE–EtOAc (5:1)] to give a white solid; yield: 0.642 g (77%); mp 189–190 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.24–7.31 (m, 2 H), 7.37 (d, J = 7.4 Hz, 1 H), 7.67 (d, J = 7.8 Hz, 1 H), 13.76 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 112.5, 121.8, 124.3 127.2, 129.4, 141.3, 189.9.

ESI-MS: m/z = 167.8 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_7H_5NS_2$: 167.9936; found: 167.9938.

6-Bromo-1,3-benzothiazole-2(3*H*)-thione (1b)
White solid; yield: 0.60 g (49%); mp 265–266 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.22 (d, J = 8.1 Hz, 1 H), 7.54 (d, J = 8.8 Hz, 1 H), 7.95 (s, 1 H), 13.86 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 113.9, 116.5, 124.2, 130.1, 131.5, 140.6, 190.1.

ESI-MS: m/z = 246.5 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_7H_4BrNS_2$: 245.9041; found: 245.9040.

4-Bromo-1,3-benzothiazole-2(3*H*)-thione (1c)
White solid; yield: 0.72 g (59%); mp 184–185 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.20 (m, 1 H), 7.57 (d, J = 8.2 Hz, 1 H), 7.67 (d, J = 7.8 Hz, 1 H), 13.73 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 103.7, 121.0, 125.5, 130.5, 131.9, 140.2, 190.9.

ESI-MS: m/z = 246.8 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_7H_4BrNS_2$: 245.9041; found: 245.9042.

6-Fluorobenzothiazole-2(3*H*)-thione (1d)
White solid; yield: 0.75 mg (81%); mp 216–217 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.21–7.32 (m, 2 H), 7.64 (dd, J = 8.2 Hz, J = 1.8 Hz, 1 H), 13.82 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 109.0 (d, $J_{F–C}$ = 28.0 Hz), 113.5 (d, $J_{F–C}$ = 9.3 Hz), 114.8 (d, $J_{F–C}$ = 24.4 Hz), 130.8 (d, $J_{F–C}$ = 10.8 Hz), 138.1, 159.2 (d, $J_{F–C}$ = 239.5 Hz), 190.0.

ESI-MS: m/z = 186.7 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_7H_4FNS_2$: 185.9842; found: 185.9844.

Methyl 2-Thioxo-2,3-dihydro-1,3-benzothiazole-6-carboxylate (1e)
White solid; yield: 0.85 g (76%); mp 265–266 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 3.86 (s, 3 H), 7.37 (d, J = 8.7 Hz, 1 H), 7.95 (d, J = 8.7 Hz, 1 H), 8.32 (s, 1 H), 13.92 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 100 MHz): δ = 52.3, 112.2, 123.3, 125.3, 128.4, 129.8, 144.8, 165.6, 191.9.

ESI-MS: m/z = 226.1 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_9H_7NO_2S_2$: 225.9991; found: 225.9987.

2-Thioxo-2,3-dihydro-1,3-benzothiazole-6-carbonitrile (1f)
White solid; yield: 0.54 g (57%); mp 272–273 °C.

^{1}H NMR (DMSO-d_6, 600 MHz): δ = 7.39 (d, J = 8.2 Hz, 1 H), 7.80 (d, J = 8.2 Hz, 1 H), 8.20 (s, 1 H), 13.94 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 150 MHz): δ = 106.4, 113.0, 118.6, 126.0, 130.3, 131.2, 144.5, 191.7.

ESI-MS: m/z = 193.1 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_8H_4N_2S_2$: 192.9889; found: 192.9893.

6-(Trifluoromethoxy)-1,3-benzothiazole-2(3*H*)-thione (1g)
White solid; yield: 0.94 g (75%); mp 234–235 °C.

^{1}H NMR (DMSO-d_6, 400 MHz): δ = 7.34–7.40 (m, 2 H), 7.83 (s, 1 H), 13.90 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 100 MHz): δ = 113.3, 115.2, 120.1 (d, $J_{F–C}$ = 191.2 Hz), 120.6, 130.8, 140.4, 144.7, 190.9.

ESI-MS: m/z = 275.1 [M + Na].

HRMS: m/z [M + H]$^+$ calcd for $C_8H_4F_3NOS_2$: 251.9759; found: 251.9761.

6-Methyl-1,3-benzothiazole-2(3*H*)-thione (1h)
White solid; yield: 0.64 g (71%); mp 178–179 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 2.32 (s, 3 H, CH_3), 7.20 (s, 2 H), 7.44 (s, 1 H), 13.68 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 20.8, 112.2, 121.6, 128.1, 129.5, 133.9, 139.2, 189.2.

ESI-MS: m/z = 205.1 [M + Na].

HRMS: m/z [M + H]$^+$ calcd for $C_8H_7NS_2$: 182.0093; found: 182.0096.

4-Bromo-6-methyl-1,3-benzothiazole-2(3*H*)-thione (1i)
White solid; yield: 0.75 g (58%); mp 219–220 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 2.31 (s, 3 H), 7.42 (s, 1 H), 7.48 (s, 1 H), 13.71 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 20.3 (s, CH_3), 103.3, 121.0, 130.5, 131.1, 135.6, 138.1, 190.4.

ESI-MS: m/z = 260.2 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_8H_6BrNS_2$: 259.9198; found: 259.9193.

6-Chloro-4-fluoro-1,3-benzothiazole-2(3*H*)-thione (1j)
White solid; yield: 0.88 g (81%); mp 210–211 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.48–7.71 (m, 1 H), 7.71–7.90 (m, 1 H), 14.12 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 114.1 (d, $J_{F–C}$ = 20.8 Hz), 117.7, 120.7, 128.7 (d, $J_{F–C}$ = 8.6 Hz), 129.7, 133.1 (d, $J_{F–C}$ = 65.2 Hz), 190.8.

ESI-MS: m/z = 219.1 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_7H_3ClFNS_2$: 219.9452; found: 219.9456.

4,6-Difluoro-1,3-benzothiazole-2(3*H*)-thione (1k)
White solid; yield: 0.81 g (80%); mp 185–186 °C.

^{1}H NMR (DMSO-d_6, 300 MHz): δ = 7.34–7.65 (m, 2 H), 13.95 (br s, 1 H).

^{13}C NMR (DMSO-d_6, 75 MHz): δ = 102.9 (d, $J_{F–C}$ = 22.2 Hz), 105.0 (d, $J_{F–C}$ = 27.2 Hz), 108.2 (d, $J_{F–C}$ = 27.2 Hz), 118.0 (d, $J_{F–C}$ = 27.2 Hz), 134.6 (d, $J_{F–C}$ = 459.6 Hz), 158.6 (d, $J_{F–C}$ = 243.8 Hz), 190.6.

ESI-MS: m/z = 203.2 [M + H].

HRMS: m/z [M + H]$^+$ calcd for $C_8H_2F_2NS_2$: 203.9748; found: 203.9750.

3-Methyl-1,3-benzothiazole-2(3*H*)-thione (1l)
White solid; yield: 0.63 g (70%); mp 95–97 °C.

^{1}H NMR ($CDCl_3$, 300 MHz): δ = 3.84 (s, 3 H), 7.20 (d, *J* = 8.3 Hz, 1 H), 7.27–7.32 (m, 1 H), 7.39–7.48 (m, 2 H).

^{13}C NMR ($CDCl_3$, 75 MHz): δ = 33.2, 112.4, 121.3, 124.9, 127.0, 127.5, 142.0, 189.4.

ESI-MS: *m/z* = 182.2 [M + H].

HRMS: *m/z* [M + H]$^+$ calcd for $C_8H_7NS_2$: 182.0093; found: 182.0091.

3-Ethyl-1,3-benzothiazole-2(3*H*)-thione (1m)
White solid; yield: 0.63 g (65%); mp 82–84 °C.

^{1}H NMR ($CDCl_3$, 300 MHz): δ = 1.40 (t, *J* = 7.2 Hz, 3 H), 4.50 (q, *J* = 7.2 Hz, 2 H), 7.23 (d, *J* = 8.3 Hz, 1 H), 7.28–7.33 (m, 1 H), 7.40–7.45 (m, 1 H), 7.50 (d, *J* = 7.9 Hz, 1 H).

^{13}C NMR ($CDCl_3$, 75 MHz,): δ = 12.0, 41.5 112.4, 121.6 124.8, 127.1, 128.1, 141.2, 188.7.

ESI-MS: *m/z* = 196.4 [M + H].

HRMS: *m/z* [M + H]$^+$ calcd for $C_9H_9NS_2$: 196.0249; found: 196.0251.

3,6-Dimethyl-1,3-benzothiazole-2(3*H*)-thione (1n)
White solid; yield: 0.72 g (74%); mp 126–128 °C.

^{1}H NMR ($CDCl_3$, 300 MHz,): δ = 2.41 (s, 3 H), 3.82 (s, 3 H), 7.08 (d, *J* = 8.3 Hz, 1 H), 7.22 (d, *J* = 8.3 Hz, 1 H), 7.27 (s, 1 H).

^{13}C NMR ($CDCl_3$, 75 MHz): δ = 21.2, 33.2, 112.1, 121.4, 127.6, 128.1, 135.1, 140.1, 188.8.

ESI-MS: *m/z* = 196.4 [M + H].

HRMS: *m/z* [M + H]$^+$ calcd for $C_9H_9NS_2$: 196.0249; found: 196.0247.

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References

(1) These compounds can exist in two tautomeric forms: the 1,3-benzothiazole-2(3*H*)-thione or the 1,3-benzothiazole-2-thiol. X-ray determination and calculation suggest that the 1,3-benzothiazole-2(3*H*)-thione tautomer is generally favored; see: (a) Curtiss, L. A.; Redfern, P. C.; Raghavachari, K.; Pople, J. A. *J. Chem. Phys.* **2001**, *114*, 108. (b) Steudel, R.; Steudel, Y. *Chem. Eur. J.* **2006**, *12*, 8589. (c) Radha, A. *Z. Kristallogr.* **1985**, *171*, 225.

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