HETEROCYCLES, Vol. 63, No. 5, 2004, pp. 1153 - 1163
Received, 1st December, 2003, Accepted, 16th February, 2004, Published online, 5th March, 2004

A CONVENIENT SYNTHESIS OF SOME NEW 1, 3, 4-BENZOTHIADIAZEPIN-5-ONES

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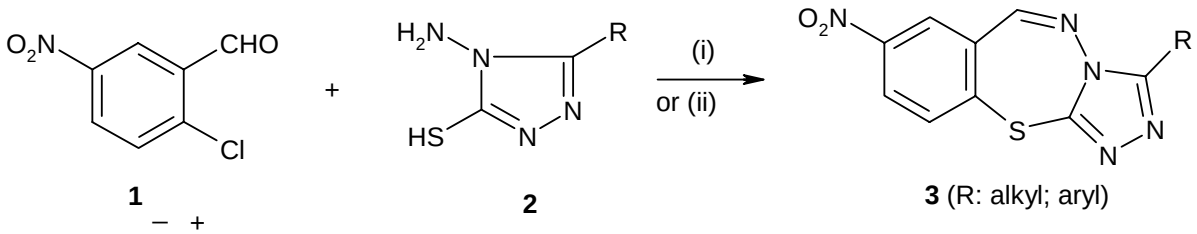
Abstract – A synthesis of new 1,3,4-benzothiadiazepin-5-ones (**6a-c**) has been achieved *via* cyclocondensation, promoted by acetic anhydride at reflux, of the corresponding acyclic 2-{[2-oxo-1-(*N*-arylhydrazono)propan-1-yl]mercapto}benzoic acids (**4a-c**). Structural assignments of the heterocyclic products (**6a-c**) are based on analytical, spectral (IR, MS and NMR) and X-Ray crystal structure data.

INTRODUCTION

The preparation and biological aspects of various isomeric benzothiadiazepines have been tackled.[1-9] These bicyclic systems include [1,2,4-],[1] [1,2,5-],[2,3] [1,4,5-],[4] [2,1,4-],[5] [2,1,5-],[6] [2,3,5-],[7] [3,2,4-][8] and [4,1,3-][9] benzothiadiazepine *S*,*S*-dioxides. The 1,3,4-benzothiadiazepine ring system, is described as condensed with heterocycles,[10-13] such as in 1,2,4-triazolo[3,4-*b*][1,3,4]benzothiadiazepines (**3**).[10,11] Derivatives of this *tricyclic* system (**3**) were prepared from 5-substituted 4-amino-3-mercapto-1,2,4-triazoles (**2**)[10,11] in which the functional appendages were utilized in constructing the 1,3,4-benzothiadiazepine upon interaction with 2-chloro-5-nitrobenzaldehyde(**1**) (Scheme 1). More recently, some derivatives of the related *tetracyclic* benzopyrano-1,2,4-triazolo[3,4-*b*][1,3,4]benzothiadiazepines were likewise prepared in a one-pot reaction of **2** with appropriately substituted flavanones.[12] However, the parent *bicyclic* 1,3,4-benzothiadiazepine system is, to our knowledge, hitherto undescribed, and for which a facile synthetic approach appears desirable.

Scheme 1

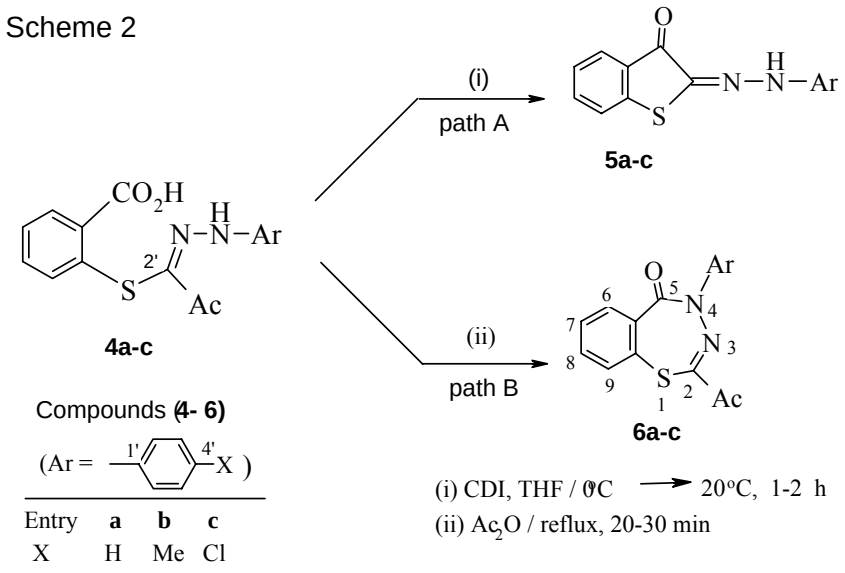


1 **2** **3** (R: alkyl; aryl)

(i) EtO⁻ Na⁺ (1 eq.) / EtOH, △ (ii) toluene + piperazine (catalyst) / △

Quite recently, we have reported on a unique transformation of 2-[(2-oxo-1-arylhydrazonopropan-1-yl)mercapto]benzoic acids (**4a-c**) into 2-(*N*-arylhydrazono)-1-benzothiophen-3-ones (**5a-c**), induced by 1,1'-carbonyldiimidazole(CDI)[13] (Scheme 2 / path A). The present work describes an alternative mode of cyclization of **4a-c**, promoted by acetic anhydride as the condensing agents, leading to the respective target 1,3,4-benzothiadiazepin-5-(4*H*)-ones (**6a-c**) (Scheme 2 / path B).

Scheme 2



RESULTS AND DISCUSSION

CHEMISTRY

The hydrazone moiety in the acyclic adducts (**4a-c**) acts as a binucleophile whereby the C-2' and the NH-atoms represent the possible nucleophilic centers. Direct interaction of these acyclic adducts namely, 2-[(2-oxo-1-arylhydrazonopropan-1-yl)mercapto]benzoic acids (**4a-c**), with acetic anhydride at reflux temperature, yielded the respective 1,3,4-benzothiadiazepin-5(4*H*)-ones (**6a-c**) (path B / Scheme 2). The formation of the latter bicyclic system implies that **4a-c** underwent cyclization involving the nucleophilic hydrazone NH- and the proximal activated carboxyl group (probably as mixed anhydride with acetic anhydride). Under these conditions the *alpha* C-2' , appended to the "S" atom, did not participate in the cyclization step as it did when CDI was used (path A).[13]

SPECTRAL DATA

The new compounds (**6a-c**) were characterized by elemental analyses, IR, MS and NMR spectral data. These data, detailed in the EXPERIMENTAL part, are consistent with the assigned structures. Thus, the measured high resolution mass (HRMS) spectral data for M^+ are in good agreement with the calculated values as suggested by their molecular formulas. DEPT and 2D (COSY, HMQC and HMBC) experiments showed correlations that helped in the ^{1}H- and ^{13}C- signal assignments to the various hydrogens and carbons. Thus, long range correlation is observed between H-9 and C–5a, the acyl CH_3 and C-2 as well as between H-6 and C-9a / C-5 in HMBC experiments for compounds (**6a-c**).

The present study also deals with structural determination of the 1,3,4-benzothiadiazepin-5-one ring system by X-Ray crystal structure measurements for **6a**, for which relevant crystallographic data are given in Tables 1 and 2. The molecular structure of **6a**, based on crystallographic data, is displayed in Figure 1. Calculations, based on X-Ray data, relating to the plane of the thiadiazepinone ring, show that the consecutive atoms S(1) – C(9A) – C(5A) – C(5) are lying quite well in the same plane, and are planar with the benzo-fused ring, whereas the other three complementary atoms C(2) – N(3) – N(4) are folded away, out of plane in a "boat" like manner (Figure 2). The phenyl ring (C(12)-C(17)) is rotated out of this plane by –63.1º (N(3)-N(4)-C(12)-C(13)) in order to keep the phenyl hydrogen atoms H13 and H17 at reasonable distances from N3 and O1, respectively (H(13)…N(3) 2.82 Å, H(17))…O(1) 2.74 Å). In the solid state, the molecules of **6a** are cross-linked by a prominent network of intermolecular hydrogen bonding [Figures 3(a)-(c)]. A dominant feature is the formation of centosymmetric dimers *via* H(8)…O(2) and H(8`)…O(2`) (D = 3.236 Å, d = 2.501 Å, Θ = 155.8 º), as well as H(9).. O(2) (D = 3.448 Å, d = 2.95 Å, Θ = 147 º)[14] (Figure 3a). The dimers are further linked in the same plane by O(1)…H(16) bonds (D = 3.446 Å, d = 2.57 Å, Θ = 154.7 º) which demonstrate once more the importance of secondary or weak C-H…O hydrogen bonds for the packing of oxygen-containing heterocyclic systems.

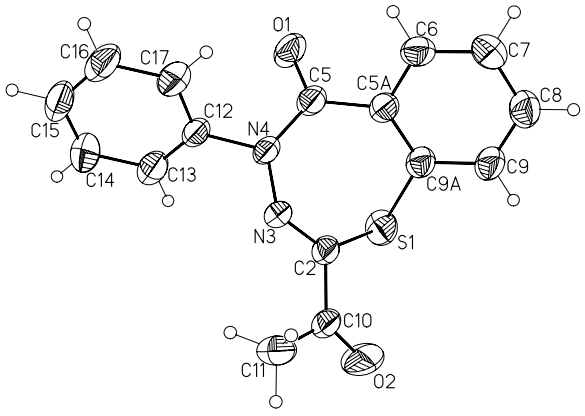


Figure 1. Thermal ellipsoid plot (50 %) of the molecular structure of **6a**

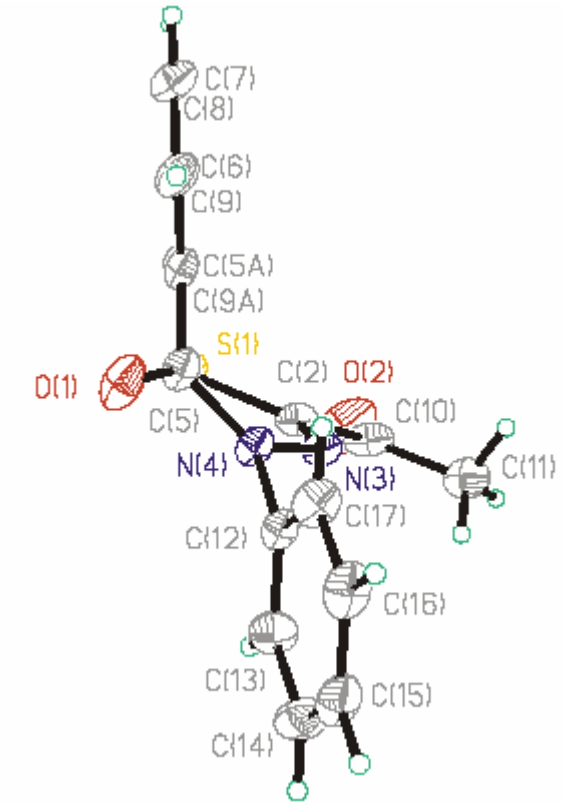


Figure 2. Thermal ellipsoid plot (50%) of the plane along the atoms S(1)-C(9A)-C(5A)-C(5) of **6a**

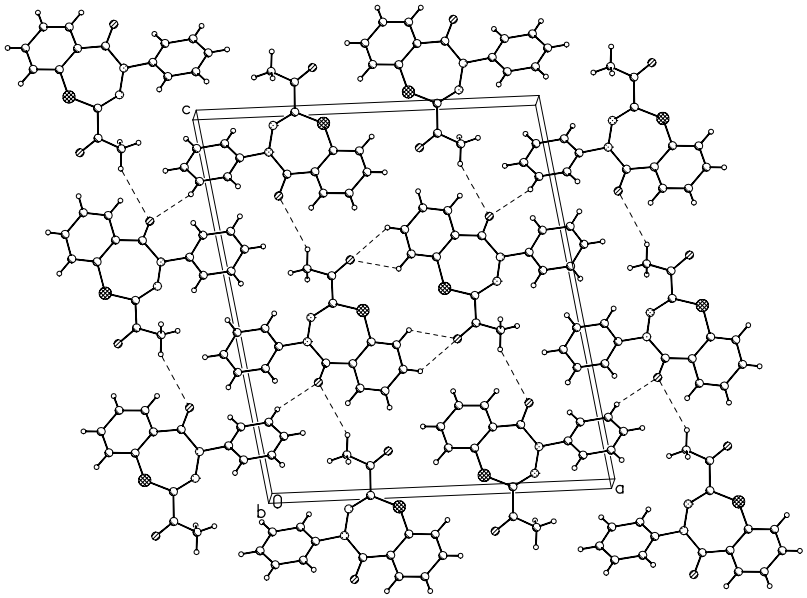


Figure 3a. Packing diagram for **6a** in the unit cell (Z = 4) showing hydrogen bonding (-viewed along the c- axis-)

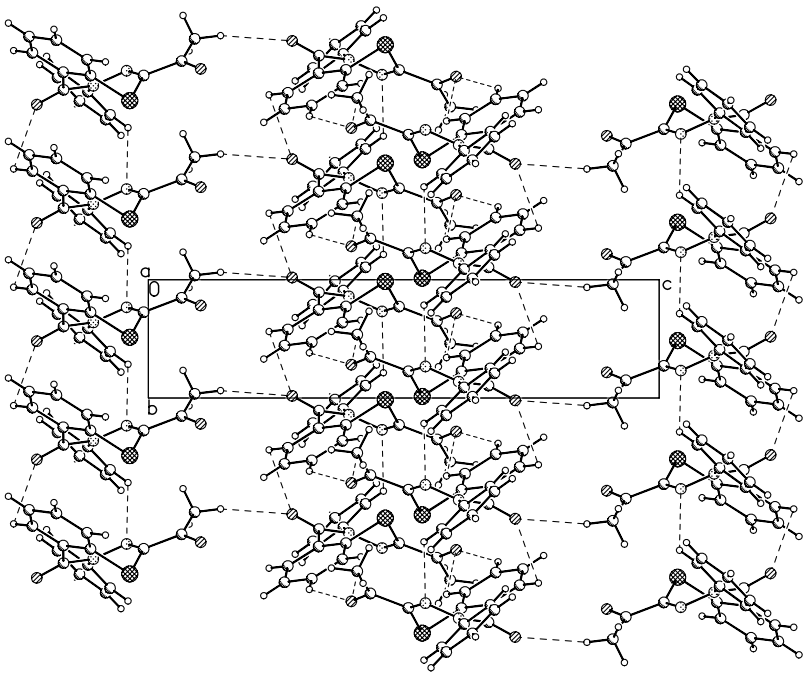


Figure 3b. Packing diagram for **6a** in the unit cell (Z = 4) showing hydrogen bonding (-viewed along the b- axis-)

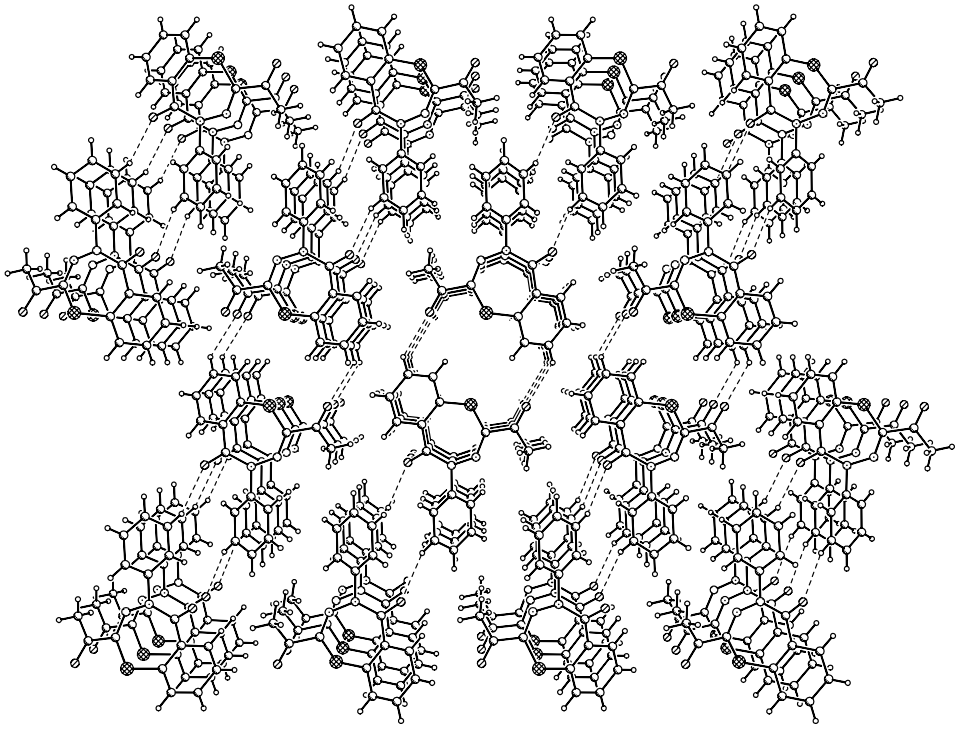


Figure 3c. Network diagram for the molecules of **6a**

Table 1. Summary of the crystal data and structure refinement parameters for 6a

Molecular formula	$C_{16}H_{12}N_2O_2S$
Formula weight	296.34 Da
Temperature (K)	223(2)
Wavelength (Å)	0.71073
Crystal system	monoclinic
Space group	$P2_1/c$
Unit cell dimensions	
a (Å)	16.745(10)
b (Å)	4.391(3)
c (Å)	19.166(11)
β(˚)	98.416(10)
Volume ($Å^3$)	1394.1(14)
Z	4
Calculated density (mg / m^3)	1.412
Absorption coefficient (mm^{-1})	0.237
F (000)	616
Crystal size (mm)	0.36 x 0.14 x 0.08
Theta range for data collection	2.31° to 28.44°
completeness to theta = 28.44°	99.4%
Index range	-22 ≤ h ≤ 22; -5 ≤ k ≤ 5 ; -25 ≤ I ≤ 25
Reflections collected	16029
Independent reflections	3490 [R_{int} = 0.0479]
Reflections used	6762
Weight scheme	Calcd $w = 1 / [\sigma^2 (F_0)^2 + (0.0586P)^2 + 0.3244P]$ where $P = [(F_0)^2 + 2(F_c)^2] / 3$
Data / restraints / parameters	2640 / 0 / 191
Goodness-of-fit on F^2	1.038
Final *R* indices [$I < 2\sigma(I)$]	$R_1 = 0.0452$, $wR_2 = 0.1158$
R indices (all data)	$R_1 = 0.0627$, $wR_2 = 0.1261$
Largest difference peak (e. $Å^{-3}$)	0.253
Largest difference hole (e. $Å^{-3}$)	-0.201

Table 2. Selected bond lengths, angles, and torsion angles for 6a

Bond lengths (Å)		Bond angles (°)	
S(1)- C(9A)	1.7686(19)	C(2)- S(1)- C(9A)	97.33(8)
S(1)- C(2)	1.7691(18)	N(3)- C(2)- S(1)	128.05(13)
N(3)- C(2)	1.2820(19)	C(2)- N(3)- N(4)	122.68(14)
N(3)- N(4)	1.3802(18)	N(3)- N(4)- C(5)	129.77(13)
N(4)- C(5)	1.402(2)	N(4)- C(50)- C(5A)	120.32(14)
C(5)- C(5A)	1.486(2)	C(5)- C(5A)- C(9A)	124.41(16)
C(5A)-C(9A)	1.391(2)	C(5A)- C(9A)- S(1)	120.46(13)
C(2)- C(10)	1.508(3)	S(1)- C(2)- C(10)	116.32(12)
N(4)- C(12)	1.448(2)	N(3)- N(4)- C(12)	111.06(11)
C(5)- O(1)	1.214(2)	N(4)- C(5)- O(1)	118.37(15)

Torsion angles (°)	
C(9A)- S(1)- C(2)- N(3)	60.90(17)
C(9A)- S(1)- C(2)- C(10)	-126.01(14)
C(10)- C(2)- N(3)- N(4)	-172.39(14)
S(1)- C(2)- N(3)- N(4)	0.8(2)
C(2)- N(3)- N(4)- C(5)	-40.4(3)
C(2)- N(3)- N(4)- C(12)	152.13(15)
C(3)- N(4)- C(5)- C(5A)	-8.7(3)
C(12)- N(4)- C(5)- C(5A)	158.00(15)
C(3)- N(4)- C(5)- O(1)	178.95(17)
C(12)- N(4)- C(5)- O(1)	-14.3(3)
C(6)- C(5A)- C(5)- O(1)	40.8(3)
C(6)- C(5A)- C(5)- N(4)	-131.32(17)
C(2)- S(1)- C(9A)- C(9)	121.86(15)
C(5)- C(5A)- C(9A)- S(1)	-2.5(2)
C(6)- C(5A)- C(9A)- S(1)	-179.08(13)

CONCLUSION

Reagent-controlled selectivity in chemical reactions is receiving continued attention and finds wide applications in the art of synthetic design. An intersesting aspect of the cyclization reactions of **4a-c** is the selective cyclocondensation behaviours toward condensing agents. Thus, annulation of **4a-c** with acetic anhydride gives 7-membered heteroring, whereas CDI directs 5-membered heteroring formation.

EXPERIMENTAL

2-Mercaptobenzoic acid and 3-chloropentane-2,4-dione were purchased from Acros. Melting points (uncorrected) were determined on a Gallenkamp electrothermal melting-temperature apparatus. ^{1}H- and ^{13}C NMR spectra were measured on a Bruker DPX-300 instrument with Me_4Si as internal reference. EIMS mass spectra were obtained using a Finnigan MAT TSQ-70 spectrometer at 70 eV; ion source temperature = 200 ^{o}C. IR spectra were recorded as KBr discs on a Nicolet Impact-400 FT-IR spectrophotometer. Microanalyses were preformed at the Microanalytical Laboratory-Inorganic Chemistry Department, Tübingen University, Germany.

2-[(2-Oxo-1-arylhydrazonopropan-1-yl)mercpto]benzoic acids (4a-c)

The title acyclic adducts have been previously described,[13] and are prepared *via* direct interaction between 2-mercaptobenzoic acid and the appropriate *N*-arylhydrazonoyl chloride in the presence of triethylamine.

2-Acetyl-4-phenyl-1,3,4-benzothiadiazepin-5(4*H*)-one (6a)

A solution of 2-[(2-oxo-1-phenylhydrazonopropan-1-yl)mercapto]benzoic acid (**4a**)[13] (1.6 g, 5.1 mmol) in acetic anhydride (20 mL) was heated at reflux for 30 min. The resulting reaction mixture was then cooled to 10 ^{o}C, cautiously diluted with cold water (50 mL) and extracted with dichloromethane (2 x 30 mL). The combined organic extracts were washed with water (20 mL), dried ($MgSO_4$), and the solvent was evaporated. The residual product was purified by column chromatography on silica gel, eluting with dichloromethane, to give the pure title compound as pale yellow solid. Yield of **6a** = 0.88 g (58%), mp 105-106 ^{o}C (recrystallized from ethanol). *Anal*. Calcd for $C_{16}H_{12}N_2O_2S$: C, 64.85; H, 4.1; N, 9.45; S, 10.8. Found: C, 64.7; H, 4.0; N, 9.3; S, 10.6; IR (KBr): ν 3066, 2940, 2903, 1700, 1665, 1598, 1534, 1491, 1435, 1305, 1211, 1152, 1121, 1019 cm^{-1}; MS *m/z* (%): 296 (4, M^+), 254 (3), 253 (3), 226 (4), 197 (6), 192 (10), 174 (11), 147 (8), 105 (35), 91 (56), 77 (100); HRMS: Calcd for $C_{16}H_{12}N_2O_2S$: 296.06196. Found: 296.06347; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.31 (s, 3H, CH_3), 7.39 (m, 1H, H-4'), 7.42-7.51(m, 5H, H-2'/ H-6', H-3'/ H-5', H-9), 7.56(dd, 1H, J = 8.7, 7.4 Hz, H-7), 7.59 (dd, 1H, J = 7.4, 7.5 Hz, H-8), 7.78 (dd, 1H, J = 8.7, 1.2 Hz, H-6); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.5 (CH_3), 127.3 (C-2' / C-6'), 128.5 (C-4'), 129.4 (C-3' / C-5'), 129.9 (C-7), 132.1 (C-9), 132.2 (C-6), 133.5 (C-9a), 134.0 (C-8), 137.7 (C-5a), 142.9 (C-1'), 155.5 (C-2), 166.4 (C-5), 193.2 (Me-*C* = O).

2-Acetyl-4-(4'-methylphenyl)-1,3,4-benzothiadiazepin-5-(4*H*)-one (6b)

This compound was prepared from the reaction of 2-{[2-oxo-1-(4-methylphenylhydrazono)propan-1-yl]mercapto}benzoic acid (**4b**)[13] (1.8 g, 5.5 mmol) with acetic anhydride (15 mL) following the same procedure and experimental conditions described above for obtaining **6a**. Yield of **6b** = 1.1 g (64%), mp 145-146 ^{o}C (recrystallized from ethanol). *Anal*. Calcd for $C_{17}H_{14}N_2O_2S$: C, 65.8; H, 4.55; N, 9.0; S, 10.0 Found: C, 65.7; H, 4.5; N, 8.9; S, 10.2; IR (KBr): *v* 2988, 2922, 2854, 1701, 1661, 1596, 1539, 1510, 1427, 1350, 1308, 1208, 1153, 1120, 1013 cm^{-1}; MS *m/z* (%): 310 (3, M^+), 268 (2), 267 (2), 224 (11), 174 (3), 162 (3), 149 (10), 147 (4), 119 (31), 105 (22), 91 (100); HRMS: Calcd for $C_{17}H_{14}N_2O_2S$: 310.07907. Found: 310.07760; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.30 (s, 3H, CH_3), 2.33 (s, 3H, Ar-CH_3), 7.27 (d, 2H, *J* = 8.0 Hz, H-3'/ H-5'), 7.38 (d, 2H, *J* = 8.0 Hz, H-2'/ H-6'), 7.44 (d, 1H, *J* = 7.6 Hz, H-9), 7.51 (dd, 1H, *J* = 7.4, 7.6 Hz, H-7), 7.57 (dd, 1H, *J* = 7.4, 7.6 Hz, H-8), 7.77 (d, 1H, *J* = 7.6 Hz, H-6); ^{13}C NMR (75 MHz, DMSO-d_6): δ 21.2 (Ar-*C*H_3), 26.5 (CH_3), 127.2 (C-2' / C-6'), 129.8 (C-7), 129.9 (C-3' / C-5'), 132.1 (C-6), 132.2 (C-9), 133.5 (C-9a), 133.9 (C-8), 137.8 (C-5a), 138.1 (C-4'), 140.4 (C-1'), 155.3 (C-2), 166.5 (C-5), 193.3 (Me-*C*=O).

2-Acetyl-4-(4'-chlorophenyl)-1,3,4-benzothiadiazepin-5(4*H*)-one (6c)

This compound was prepared from the reaction of 2-{[2-oxo-1-(4-chlorophenylhydrazono)propan-1-yl]mercapto}benzoic acid (**4c**)[13] (1.4 g, 4.0 mmol) with acetic anhydride (15 mL) following the same procedure and experimental conditions described above for obtaining **6a**. The product was recrystallized from ethanol. Yield of **6c** = 0.94 (71%), mp 136-137 ^{o}C. *Anal*. Calcd for $C_{16}H_{11}N_2O_2ClS$: C, 68.1; H, 3.35; N, 8.5; Cl, 10.7; S, 9.7. Found: C, 57.9; H, 3.3; N, 8.4; Cl, 10.7; S, 9.55; IR (KBr): *v* 3087, 3055, 3010, 1696, 1667, 1601, 1551, 1493, 1428, 1351, 1313, 1197, 1146, 1010 cm^{-1} ; MS *m/z* (%): 330 (4, M^+), 288 (5), 260 (2), 224 (6), 197 (3), 174 (3), 162 (15), 161 (15), 139 (91), 125 (42), 111 (100), 90 (31); HRMS: Calcd for $C_{16}H_{11}N_2O_2ClS$: 330.022977. Found: 310.002565; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.31 (s, 3H, CH_3), 7.45 (d, 1H, *J* = 7.6 Hz, H-9), 7.50 (dd, 1H, *J* = 7.6, 8.2 Hz, H-7), 7.52 (d, 2H, *J* = 8.6 Hz, H-3'/ H-5'), 7.56 (d, 2H, *J* = 8.6 Hz, H-2'/ H-6'), 7.59 (m, 1H, H-8), 7.78 (dd, 1H, *J* = 7.6, 1.2, H-6); ^{13}C NMR (75 MHz, DMSO-d_6): δ 26.5 (CH_3), 129.1 (C-3' / C-5'), 129.4 (C-2' / C-6'), 129.9 (C-7), 132.2 (C-6), 132.3 (C-9), 132.9 (C-1'), 133.4 (C-9a), 134.1 (C-8), 137.4 (C-5a), 141.6 (C-4'), 156.2 (C-2), 166.4 (C-5), 193.1 (Me-*C*=O).

Collection of X-Ray Diffraction Data and the Structure Analysis of 6a

Yellow rod crystals of **6a** were grown by allowing a clear solution of **6a** in dichloromethane/methanol to evaporate slowly at room temperature over 3-4 days. Data were colleced with a Siemens SMART CCD diffractometer [Mo-Kα radiation, graphite monochromator] operating in the omega scan mode (0.3^{o}).

The data were reduced with the Siemens-Bruker program suite XSCANS, [15] and the structure was solved by the direct method using SHELXTL PLUS program.[16] All non-hydrogen atoms were refined anisotropically by full-matrix, least-squares procedure based on F^2 using all unique data. The hydrogen atoms were located from the difference Fourier electron density synthesis and were then refined isotropically using a 'riding model'.

Supplementary Material

Crysallographic data for the structural analysis have been deposited with the Cambridge Crystallographic Data Center, CCDC No. 224403 for compound (**6a**). Copies of further information may be obtained free of charge from the Director, CCDC, 12 Union Road, Cambridge CB2 1 EZ, UK (Fax: +44-1223-336033); e-mail: (deposit@ccdc.cam.ac.uk or http://www.ccdc.cam.ac.uk).

ACKNOWLEDGEMENTS

We wish to thank the Deanship of Academic Research, University of Jordan (Amman – Jordan) for financial support.

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