Contents lists available at ScienceDirect

Journal of Molecular Structure

journal homepage: www.elsevier.com/locate/molstruc

MOLECULAR

Synthesis and characterisation a series of N-(3,4-dichlorophenyl)-N'-(2,3 and 4-methylbenzoyl)thiourea derivatives

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ARTICLE INFO

Article history: Received 19 January 2010 Received in revised form 26 April 2010 Accepted 26 April 2010 Available online 29 April 2010

Keywords: Methylbenzoylthiourea Carbonylthiourea Thiourea Crystal structure

1. Introduction

ABSTRACT

Three new compounds, *N*-(3,4-dichlorophenyl)-*N*-(2-methylbenzoyl)thiourea (**2a**), *N*-(3,4-dichlorophenyl)-*N*'-(3-methylbenzoyl)thiourea (**2b**) and *N*-(3,4-dichlorophenyl)-*N*'-(4-methylbenzoyl)thiourea (**2c**) are isomers which have been successfully synthesised and characterised by typical spectroscopic techniques, namely IR and ¹H and ¹³C multi-nuclear magnetic resonance (NMR). The structure of **2a** was determined by single crystal X-ray diffraction method and crystallised in the monoclinic system with space group C2/c, adopting configuration of the molecule is *trans-cis*. The infrared spectra of these compounds showed four significant stretching vibrations of v(N-H), v(C=O), v(C-N) and v(C=S) at 3261–3279 cm⁻¹, 1662–1680 cm⁻¹, 1345–1379 cm⁻¹ and 688–738 cm⁻¹, respectively. The ¹³C NMR chemical shift for the thiourea moiety appeared at *ca*. δ_C 180 ppm.

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For the past few decades, thiourea derivatives have attracted great attention as versatile ligands in numerous applications [1]. This is due to its unique properties which enable to coordinate with various transition metal ions as monodentate or bidentate ligands [2-5]. Thiourea derivatives for instance derived as substituted benzovlthiourea or phenylthiourea derivatives are attractive model compounds for the studies in solid-state chemistry due to their tendency for the formation of intra- and intermolecular hydrogen bonds of the N-H proton-donor groups to sulphur and carbonyl oxygen atoms [7-25]. To date, these derivatives are widely used in numerous applications such as in pharmaceutical industry for potential therapeutic agents as antibacterial [26-28], anti-HIV [28,29], anticancer drugs [30–32], antidepressants [33,34] and antihyperlipidemic, antiallergic, antiparasitic, platelet antiaggregating, antiproliferative activities [35-37]. Previous studies have reported that the compounds containing thiourea moieties have been used extensively and commercially as herbicides, fungicides and insecticides agents in the agrochemical industries [23,38-41].

Several studies have revealed that thiourea derivatives not only have been using in medical and agriculture applications but also they play major contribution in the environmental and industrial applications. In the case of 1-benzoyl-3-propylthiourea, this compound has great interest as effective adsorbent for removal of mercury ions from aqueous solutions [24]. The chelating resin was

* Corresponding author. E-mail address: mohdsukeri@umt.edu.my (M.S.M. Yusof). reported derived from copolymerization of bisthiourea/thiourea/ glutaraldehyde which was successfully applied to determine Hg(II) from a real matrix [42]. On the other hand, thiourea and its derivatives can serve as useful host material or inclusion compounds exhibiting wide range applications in the development of electronics and optoelectronics devices [43–45]. Thiourea and its derivatives have been found as an effective corrosion inhibitors agent because sulphur atom is easily protonated in acidic solution [46– 48]. Nowadays, these derivatives have been employed successfully as catalyst in the palladium-catalyzed Suzuki and Heck reactions because thiourea ligands are thermally stable and insensitive to air and moisture which is the reaction may be conducted at ambient environment [49–51].

In the present study, we focus on the synthesis and characterisation of three new synthetic derivatives which are 2,3 and 4methylbenzoyl with 3,4-dichlorophenyl groups as precursors attached at the terminal of two nitrogen atoms as shown in Fig. 1. Their structures were confirmed by several spectroscopic methods namely IR, ¹H and ¹³C NMR spectroscopy and single crystal X-ray diffraction structural analysis. However, only **2a** was obtained as a single crystal and suitable for single crystal X-ray diffractometer structural analysis.

2. Experimental

2.1. Physical measurements

All reactions were carried out under an ambient atmosphere and no special precautions were taken to exclude air or moisture



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Fig. 1. The proposed structures of the *N*-(3,4-dichlorophenyl)-*N*'-(2-methylbenzoyl)thiourea, **2a**, *N*-(3,4-dichlorophenyl)-*N*'-(3-methylbenzoyl)thiourea, **2b** and *N*-(3,4-dichlorophenyl)-*N*'-(4-methylbenzoyl)thiourea, **2c**.

during work-up. All chemicals were purchased from Sigma Aldrich or MERCK and used as received without further purification. Infrared spectra of the synthesised compounds were recorded from KBr pellets using FTIR Perkin Elmer 100 Spectrophotometer in the spectral range of 4000–400 cm⁻¹. ¹H 400.11 MHz and ¹³C 100.61 MHz NMR spectra were recorded using Bruker Avance III 400 Spectrometer in DMSO-d₆ as a solvent at room temperature in the range between 0–15 ppm and 0–200 ppm. Room temperature diffraction data for 2a was collected on a Bruker SMART APEX 4 K CCD diffractometer (Mo K α radiation, λ = 0.71073 Å). The structure was solved and refined by using SHELX suit [26]. All nonhydrogen atoms were refined anisotropically. The perspective view of the molecule was obtained using ORTEP-32 for Windows [27]. Data collection: SMART [28]; cell refinement: SAINT [28]; data reduction: structure: SHELXS 97 [29]; molecular graphics: SHELXL 97 [29]; software used to prepare material for publication: SHEL-XTL [30] and PLATON: software to calculate the hydrogen bonds [52].

2.2. Preparation of N-(2-methylbenzoyl)-N'-(3,4dichlorophenyl)thiourea, **2a**

The mixture of 2-methylbenzoyl chloride (2.0 g, 13 mmol) with the equimolar amount of ammonium thiocyanate (0.98 g, 13 mmol) and 3,4-dichloroaniline (2.10 g, 13 mmol) in 85 mL acetone was put at reflux with constant stirring for ca. 4 h. The solution was poured into a beaker containing some ice blocks. Then, the resulting white precipitate was filtered and washed with little cold methanol, then dried in vacuum desiccator. Finally, the crude product was purified by flash chromatography using (silica, hexane: ethyl acetate 7:3) followed by recrystallisation from methanol to afford the title compound as colourless single crystalline solids (2.65 g, 66%). IR (KBr pellets): v(N-H) 3270.93 cm⁻¹, v(C=0)1679.16 cm⁻¹, v(C–N) 1379.21 cm⁻¹, v(C=S) 722.00 cm⁻¹. ¹H NMR (DMSO-d₆, 400.11 MHz): δ 2.44 (s, 3H, Me); 7.31 (d, $J_{\rm HH}$ = 8 Hz, 2H, C₆H₃); 7.43 (t, $J_{\rm HH}$ = 4 Hz, 1H, C₆H₄); 7.51 (d, $J_{\rm HH}$ = 7 Hz, 1H, C₆H₄); 7.67 (s, 2H, C₆H₄), 8.16 (s, 1H, C₆H₃); 11.88, 12.52 (2 \times s, 1H, NH). ¹³C NMR (DMSO-d₆, 100.61 MHz): δ 20.0 (s, Me); 125.3, 126.0, 126.5, 128.6, 128.7, 130.9, 131.1, 131.2, 131.5, 134.3, 136.6, 138.5 $(12 \times s, Ar)$; 170.9 (s, C=0); 180.0 (s, C=0); 180. C=S).

2.3. Preparation of N-(3-methylbenzoyl)-N'-(3,4dichlorophenyl)thiourea, **2b**

In the similar manner as described above, compound **2b** was obtained as colourless crystalline solids (2.88 g, 85%). IR (KBr pellets): v(N-H) 3260.55 cm⁻¹, v(C=O) 1663.55 cm⁻¹, v(C-N) 1344.93 cm⁻¹, v(C=S) 687.58 cm⁻¹. ¹H NMR (DMSO-d₆,

400.11 MHz): δ 2.40 (*s*, 3H, Me); 7.42–7.50 (*m*, 2H, C₆H₃); 7.63–7.70 (*m*, 2H, C₆H₄); 7.77 (d, *J*_{HH} = 7 Hz, 1H, C₆H₄); 7.83 (*s*, 1H, C₆H₄); 8.13 (*s*, 1H, C₆H₃); 11.64, 12.59 (2 × *s*, 1H, NH). ¹³C NMR (DMSO-d₆, 100.61 MHz): δ 21.3 (*s*, Me); 125.4, 126.4, 126.6, 128.7, 128.9, 129.6, 130.9, 131.2, 132.4, 134.3, 138.4, 138.6 (12 × *s*, Ar); 168.7 (*s*, C=O); 180.1 (*s*, C=S).

2.4. Preparation of N-(4-methylbenzoyl)-N'-(3,4chlorophenyl)thiourea, **2c**

In the similar manner as described above, the compound **2c** was obtained as colourless crystalline solids by recrystallisation from methanol (0.26 g, 70%). IR (KBr pellets): v(N-H) 3278.56 cm⁻¹, v(C=O) 1661.78 cm⁻¹, v(C-N) 1374.75 cm⁻¹, v(C=S) 738.12 cm⁻¹. ¹H NMR (DMSO-d₆, 400.11 MHz): δ 2.40 (s, 3H, Me); 7.37 (pseudo-d, $J_{HH=8}$ Hz, 2H, C_6H_4); 7.60–7.69 (m, 2H, C_6H_3); 7.76 (s, 1H, C_6H_3); 8.13 (pseudo-d, J_{HH} = 13 Hz, 2H, C_6H_4); 11.60, 12.61 (2 × s, 1H, NH). ¹³C NMR (DMSO-d₆, 100.61 MHz): δ 21.5 (s, Me); 120.7, 121.9, 126.5, 128.2, 128.7, 129.3, 129.5, 130.9, 131.0, 131.2 (10 × s, Ar); 168.4 (s, C=O); 180.2 (s, C=S).

3. Results and discussion

3.1. Spectroscopic studies

Infrared spectra of these title compounds reveal all the expected frequency region of the v(N–H), v(C=O), v(C–N) and v(C=S). The bands at 3279-3261 cm⁻¹ and 3236-3094 cm⁻¹ represent asymmetric and symmetric stretching vibration respectively of the v(N-H) in the secondary thioamide group. These assignments were supported by the literature that $v_{N(1)-H(1)}$ can be seen at above 3200 cm^{-1} and the $v_{N(2)-H(2)}$ can be found at above 3000 cm⁻¹ have been examined due to the existence of intramolecular hydrogen bonding [1-3,12-15,33,34,53-59]. The trans-cis conformation of all studied compounds is related to the N-H stretching frequencies range which are depends on the position – NHC(S)NHC(O)- group vibrations and stabilised by hydrogen bonding. The carbonyl band $v_{C=0}$ of **2a–c** are clearly observed at above 1660 cm⁻¹ which might be related to the resonance effect with the phenyl rings and existence of intramolecular hydrogen bonding with N-H [1-3,12-15,54-56]. The bands sp² of the carbonyl stretching for 2a and b favour to absorb strongly at 1680 cm⁻¹ and 1664 cm⁻¹ compared to the **2c** absorbs weakly at 1662 cm⁻¹. These vibrational frequencies occur at variation intensities in IR spectra because of the polarity of the double bond and electron donating methyl groups at ortho, meta or para position of the substituent on the aromatic. The $v_{(C-N)}$ stretching frequencies have been found at around 1345–1379 cm⁻¹. In fact, these vibrational frequencies have been assigned by comparison with the assignments of acylthiourea derivatives at 1400–1000 cm⁻¹ [3,12,15,54,56] and these assignments of thiourea derivatives were also confirmed by [34,53,57,58]. Weigun et al. reported the v(C-N)stretching frequencies at *ca*. 1400 cm⁻¹ for the *N*-2-fluorobenzoyl-N'-2-methoxyphenylthiourea [15]. Whilst, the $v_{(C-S)}$ stretching vibration can be observed at 688–738 cm⁻¹ range that are in close agreement with previously studied of other thiourea derivatives [12.34.54.56–58]. For instance, these vibrational modes show a good agreement with the 3-monosubstituted furovlthioureas series in the wide range at *ca*. 700 cm^{-1} [54]. The characteristic region of the high frequency v(C=S) in the furoylthiourea derivatives are described as large double bond character and the lower nucleophilic character of the sulphur atom in comparison with alkylthioureas. Comparing to this report, it can be concluded that the lowest frequency of the v(C=S) in the spectrum of **2b** has less double bond character between to the **2a** and **c**. On other hand, the vibrational frequencies in this range are influenced by different position of the substituent methyl groups on the aromatic and probably due to the increased steric repulsion between the bulky sulphur atom and the hydrogen in 3,4-dichlorophenyl and methylbenzoyl in aromatic nuclei. The bands observed at *ca*. 800 cm⁻¹ in the spectra corresponds to v(C-CI) stretching vibrations with the electronegativities of the meta and para chloro substituted of phenyl ring compared to those v(C-F) stretching vibrations at *ca*. 1100 cm⁻¹ in the *N*-2-fluorobenzoyl-*N*'-4-methoxyphenylthiourea [12] with bearing substituent effect in ortho fluoro group on the aromatic. The FTIR spectra are shown in Fig. 2, respectively to its designated molecule. In ¹H NMR spectra of the **2a-c**, one resonance can be observed at $\delta_{\rm H}$ 2.40–2.44 ppm as singlet which are due to methyl proton substituents attached to the phenyl ring. The unresolved resonance of aromatic protons of 2a-c can be clearly observed as distinctive multiplet resonances between $\delta_{\rm H}$ 7.31-8.16 ppm. These characteristics are strongly influenced by the o, *m* and *p*-substituent methyl groups at the phenyl ring and due to the overlapping proton signals in the aromatic rings. Two signals noted as singlet, corresponds to one proton at *ca*. $\delta_{\rm H}$ 11 ppm which is assigned to the N(1)H proton which is substituted to the carbonyl and N(2)H proton and to the thione presence at *ca*. $\delta_{\rm H}$ 12 ppm. These signals are different in term of chemical shift which is due to the intramolecular hydrogen bonded N-H bonds in the *trans* and *cis* conformations, respectively. As seen both NH signals are very close chemical shifts between $\delta_{\rm H}$ 11–12 ppm due to deshielding aromatic ring. In ¹³C NMR spectra, signals of the methyl carbon can be expected at above δ_C 20 ppm. Meanwhile, the aromatic carbon resonances can be found in between δ_{C} 120.7-138.6 ppm which is corresponding to phenyl rings in the compounds. The carbonyl and thione carbons can be clearly observed at between δ_C 168.7–180.2 ppm and more slightly deshielded in the spectra. Formation of intramolecular hydrogen bonding, the increasing electronegatively of oxygen and sulphur atoms and different environment and conformations cause a deshielding effect for these signals.

3.2. Crystal structure determination of 2a

Molecule **2a** is discrete, crystallised in monoclinic crystal system with C2/c space group, unit cells a = 10.888(3) Å, b = 10.533(3) Å, c = 28.131(7) Å and $\beta = 100.803(5)^\circ$. The crystallographic and refinement data of **2a** is shown in Table 1. The title

Table 1	1
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Parameter	Data
Empirical formula	$C_{15}H_{12}Cl_2N_2OS$
Formula weight	339.23
Crystal system	Monoclinic
Space group	C2/c
Unit cell dimensions	$a = 10.888(3)$ Å, $\alpha = 90^{\circ}$
	$b = 10.533(3)$ Å, $\beta = 100.803(5)^{\circ}$
	$c = 28.131(7)$ Å, $\gamma = 90^{\circ}$
Volume	3168.9(13) A ³
Z, Calculated density	8, 1.422 Mg/m ³
F(000)	1392
Crystal size	$0.50\times0.33\times0.17$
Theta range for data collection	2.71–28.35°
Limiting indices	−14 <= h <= 13
	<i>−</i> 14 <= <i>k</i> <= 14
	-37 <= 1 <= 34
Reflections collected/unique	11,536/3928 [R(int) = 0.0237]
Completeness to theta = 26.00	99.5%
Max. and min. transmission	0.9138 and 0.7740
Refinement method	Full-matrix least-squares on F^2
Data/restraints/parameters	3928/0/198
Goodness-of-fit on F ²	1.042
Final R indices [I > 2 sigma(I)]	R1 = 0.0544, wR2 = 0.1299
R indices (all data)	<i>R</i> 1 = 0.0756, <i>wR</i> 2 = 0.1404
Largest diff. peak and hole	0.322 and -0.192 eA ⁻³

compound, **2a** is analogous to the previously reported, 1-benzoyl-3-(3,4-dichlorophenyl)thiourea [20], N-(3,4-dichlorophenyl)-N'-(phenylacetyl)thiourea [21] and N-(butanoyl)-N'-(3,4-dichlorophenyl) thiourea [22] with the difference of 2-methylphenyl is replaced by the benzoyl, butanoyl and phenylacetyl groups (Fig. 3). The compound adopts *trans-cis* configuration with respect to the position of the 2-methylphenyl and 3,4-dichlorophenyl group relative to the S1 atom across their C9–N1 and C9–N2 bonds, respectively (Fig. 3), comparable to those other methylbenzoylthiourea derivatives reported [18–19,23–24].

The bond length and angles (Table 2) are similar and in the range of previously reported [35–37]. The bond lengths N2–C9 (1.329(3) Å) and N1–C8 (1.377(3) Å) are shorter than the normal C–N (1.472 Å) bond length indicate of partial double bond character [12]. These can be attributed to the presence of resonance in this fragment agreement with the literature [6,12] and forming a planar six-membered ring with N2–H2…O1 hydrogen bond. The



Fig. 2. IR spectra of all the synthesised compounds.



Fig. 3. The ORTEP diagram of **2a**, showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level. The dashed line indicates hydrogen bonds.

Table 1	2
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Selected bond lengths (Å) and angles (°) for 2a.

Bond lengths (Å)			
S1-C9 1.667(2)		Cl1-C13	1.730(2)
O1-C8 1.216(3)		Cl2-C14	1.727(3)
N1-C8 1.377(3)			
N1-C9 1.392(3)			
N2-C9 1.329(3)			
Bond angles (°)			
01-C8-C6	123.9(2)	C8-N1-C9	128.55(19)
N1-C8-C6	113.28(19)	C9-N2-C10	126.65(19)
N2-C9-N1	116.11(19)	C12-C13-Cl1	118.9(2)
N2-C9-S1	125.69(17)	C15-C14-Cl2	118.4(2)
N1-C9-S1	118.19(16)	C13-C14-Cl2	121.11(18)

central thiourea moiety (S1/N1/N2/C9), 2-methylphenyl (C1–C7), and 3,4-dichlorophenyl (C10–C15/Cl1/Cl2) groups are essentially planar with maximum deviation of 0.027(2) Å for atom N2 from the least-squares plane. The central thiourea moiety makes dihedral angles with the 2-methylphenyl and 3,4-dichlorophenyl fragments of 59.72(9)° and 47.38(7)°, respectively. Plane of 2methylphenyl and 3,4-dichlorophenyl group are inclined to each other at an angle of 12.63(8)°.

Table 3

Hydrogen bond geometry (Å, $^\circ).$

D–H···A	D-H	$H{\cdot}{\cdot}{\cdot}A$	$D{\cdots}A$	$D{-}H{\cdots}A$
N1-H1A···S1 ⁱ N2-H2A···O1	0.91(3) 0.94(3)	2.66(3) 1.88(3)	3.559(2) 2.663(3)	166(3) 140(3)
N2-H2A···O1 ⁱⁱ	0.94(3)	2.47(3)	3.101(3)	350(4)

Symmetry codes: (i) -1/2 - x, 1/2 - y, 1 - z; (ii) 1/2 - x, 1/2 - y, 1 - z.



Fig. 4. Packing diagram of N-(3,4-dichlorophenyl)-N-(2-methylbenzoyl)thiourea, **2a**, viewed down the *c* axis. The dashed lines denote intermolecular hydrogen bonds.

There is an intramolecular hydrogen bond, N2–H2A···O1 closing a pseudo-six-membered ring (O1···H2A–N2–C9–N1–C8–O1). In the crystal lattice, the molecules are linked by intermolecular hydrogen bonds N1–H1A···S1 and N2–H2A···O1 (Table 3), forming one-dimensional network along the *a*-axis (Fig. 4).

4. Conclusion

The derivatives of methylbenzoylthiourea which are N-(3,4-dichlorophenyl)-N'-(2-methylbenzoyl)thiourea, **2a**, N-(3,4-dichlorophenyl)-N'-(3-methylbenzoyl)thiourea, **2b** and N-(3,4-dichlorophenyl)-N'-(4-methylbenzoyl)thiourea, **2c** have been successfully prepared and fully characterised by typical spectroscopic methods. The molecular structure of N-(3,4-dichlorophenyl)-N'-(2-methylbenzoyl)thiourea, **2a** was determined using single crystal X-ray diffractometer and exhibit monoclinic crystal system. Their conformation is analogous to other benzoylthioureas and the presence of inter- and intramolecular hydrogen bonding are supported by the IR spectra, NMR measurements and single crystal X-ray diffractometer data. These derivatives are currently bring great potential and interest in the coordination chemistry as polydentate ligands to bind with wide range of metal ions for numerous promising applications in the near future.

5. Supplementary material

Crystallography has been deposited with the Cambridge Crystallography Data Centre, 12 Union road, Cambridge CB22 1EZ, UK (fax: +44 1223 336 033; email: deposit@ccdc.cam.ac.uk or http:// www.ccdc.cam.ac.uk) and is available on request quoting the deposition number CCDC 761512.

Acknowledgement

The authors gratefully acknowledge Ministry of Higher Education Malaysia for FRGS research Grant No: 59001, Ministry of Science Technology and Innovation (MOSTI), HRD (S&T) National Science Fellowship (NSF) for the postgraduate scholarship, Universiti Malaysia Terengganu and Universiti Kebangsaan Malaysia for providing research facilities.

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