Synthesis of Some New Benzimidazolyl Thioxoacetamide Derivatives

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ABSTRACT

Treatment of 2-aminobenzimidazole **1** with chloroacetyl chloride **2** in dry benzene gave N-(1*H*-1,3-benzimidazol-2-yl)-2-chloroacetamide **3**. Further reaction of **3** with a mixture of elemental sulfur and corresponding amine in dimethylformamide (DMF) under room temperature afforded benzimidazolyl thioxoacetamide **4a-n**, which recrystallized from ethanol. ^{1}H-NMR, ^{13}C-NMR and IR spectroscopy, and elemental analysis were used for identification of these compounds.

Keywords: Benzimidazole, thioxoacetamide, sulfur, amine

INTRODUCTION

Thio-oxamides have been extensively studied and found to have diverse chemical reactivity and broad spectrum of biological activity (1-3). Several of them have shown inhibiting effect on certain bacteria and dehydrogenases (3). Thio-oxamides have remarkable characteristic including thermal stability, high sublimation temperature due to having strong hydrogen bonding (4). They are also form an important group of ligands in coordination chemistry (5). It is well established that heterocycles containing a benzimidazole moiety exhibit a large variety of biological activities including antihepatitis C virus (HCV), antiviral, antibacterial and antifungal activities (6,7). Therefore, synthesis of thio-oxamides containing a benzimidazole ring still remains an active research area. Although, several thio-oxamides have been prepared (8-13) due to incessant interest in the chemistry of thio-oxamides, substantial attention should be paid to the synthesis of novel thio-oxamide derivatives especially those containing benzimidazole and amine groups.

In view of these reports and in line with our researches program in synthesis of heterocycles (14) especially those having a benzimidazole moiety (15-18), we are going to report the synthesis of some novel benzimidazolyl thioxoacetamide derivatives.

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EXPERIMENTAL

All used chemicals were prepared from Merck or Fluka Company. Melting points were determined on an electrothermal digital melting point apparatus. The IR spectra were recorded on Unicom Galaxy series FT-IR 5000 spectrometer. NMR spectra were recorded on a Bruker (300 MHz) spectrometer. Chemical shifts (ppm) were referenced to the internal standards tetramethylsilane (TMS). Microanalyses were performed by the Elemental Analyzer (Elemental, Vario EL III). The Microanalyses results were agreed favorably with the calculated values. Reactions were monitored by thin layer chromatography using silica gel F_{254} aluminum sheets (Merck).

Preparation of *N*-(1*H*-benzimidazol-2-yl)-2-chloroacetamide (3)

Chloroacetyl chloride (6 mmol) was slowly added to a solution of 2-aminobenzimidazole (3 mmol) in dry benzene which kept at to 0-5 °C. The reaction mixture was refluxed for 4 h and the excess solvent removed under vacuum. The result was washed with 5% solution of $NaHCO_3$ and then water. The crude product was recrystallized from ethanol to give white pure compound. Yield 85%; m.p. 208-210 °C; IR (KBr, cm^{-1}): 3338 (NH Stretching of amine), 3219 (NH Stretching of amine), 3080 (CH Stretching of aromatic ring), 2966 (CH Stretching of aliphatic), 1687 (C=O Stretching of amide group), 1577 (C=C Stretching of aromatic ring), 744 (C-Cl). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.08 (2H, s, NH), 7.09-7.46 (4H, m, aromatic), 4.37 (2H, s, CH_2). ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 43.9, 114.3, 121.9, 135.7, 147.4, 167.7; Anal Calcd for: $C_9H_8N_3OCl$, C, 51.06; H, 3.81; N, 20.00. Found: C, 51.34; H, 4.02; N, 19.97.

Preparation of benzimidazolyl thioxoacetamides (4)

A mixture of the appropriate amine (1.04 mmol) and sulfur (4.15 mmol) in DMF (5 mL) was stirred at room temperature for 1 h. *N*-(1*H*-benzimidazol-2-yl)-2-chloroacetamide **3** (9.54 mmol) was slowly added and the stirring continued at room temperature for a further 2-3 days. The completion of reaction was followed by TLC. After completion of reaction, water was added to precipitate the crude product, which then filtered and washed with water/acetone. The crude product was recrystallized from a mixture of EtOH/H_2O to give the pure product.

***N*-(1*H*-benzimidazol-2-yl)-2-(propylamino)-2-thioxoacetamide (4a)**

Light yellow crystals. Yield 56%; m.p. 181-183 °C; IR (KBr, cm^{-1}): 3300 (NH Stretching of amine), 3265 (NH Stretching of amine), 3050 (CH Stretching of aromatic), 2958 (CH Stretching of aliphatic), 1682 (C=O), 1564 (C=C Stretching of aromatic ring), 1265 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.05 (2H, s, 2NH), 10.89 (1H, s, NH), 7.14-7.42 (4H, m, aromatic), 3.52-3.59 (2H, m, CH_2), 1.59-1.72 (2H, m, CH_2), 0.94 (3H, t, CH_3, J = 7.4 Hz); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 11.9, 20.7, 47.3, 86.4, 114.2, 122.4, 134.9, 147.6, 163.2; Anal Calcd for: $C_{12}H_{14}N_4OS$, C, 54.94; H, 5.38; N, 21.36; S, 12.22. Found: C, 54.83; H, 5.33; N, 21.54; S, 12.36.

***N*-(1*H*-benzimidazol-2-yl)-2-(butylylamino)-2-thioxoacetamide (4b)**

Green crystals; Yield 53%; m.p. 191-193 °C; IR (KBr, cm^{-1}): 331 (NH Stretching of amine), 3273 (NH Stretching of amine), 3013 (CH Stretching of aromatic ring), 2957 (CH Stretching of aliphatic), 1672 (C=O), 1570 (C=C Stretching of aromatic ring), 1271 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.15 (2H, s, 2NH), 10.87 (1H, s, NH), 7.07-7.68 (4H, m, aromatic), 3.56-3.63 (2H, m, CH_2), 1.57-1.67 (2H, m, CH_2), 1.28-1.41 (2H, m, CH_2), 0.93 (3H, t, CH_3, J = 7.3 Hz); Anal Calcd for: $C_{13}H_{16}N_4OS$, C, 56.50; H, 5.84; N, 20.27; S, 11.60. Found: C, 56.74; H, 5.58; N, 20.01; S, 11.48.

N-(1H-benzimidazol-2-yl)-2-(sec-butylamino)-2-thioxoacetamide (4c)

Green crystals; Yield 60%; m.p. 181-183 °C; IR (KBr, cm^{-1}): 3257 (NH Stretching of amine), 3016 (CH Stretching of aromatic ring), 2964 (CH Stretching of aliphatic), 1683 (C=O), 1570 (C=C), 1273 (C=S). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.08 (2H, s, 2NH), 10.65 (1H, s, NH), 7.14-7.48 (4H, m, aromatic), 4.35-4.39 (H, m, CH), 1.56-1.71 (2H, m, CH_2), 1.21 (3H, d, CH_3, J = 6.1 Hz), 0.91 (3H, t, CH_3, J = 7.0 Hz,); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 10.9, 18.5, 27.8, 52.8, 114.0, 122.3, 134.8, 147.9, 164.2, 189.3; Anal Calcd for: $C_{13}H_{16}N_4OS$, C, 56.50; H, 5.84; N, 20.27; S, 11.60. Found: C, 56.41; H, 5. 70; N, 20.45; S, 11.80.

N-(1H-benzimidazol-2-yl)-2-(hexylamino)-2-thioxoacetamide (4d)

Green crystals; Yield 61%; m.p. 184-186 °C; IR (KBr, cm^{-1}): 3265 (NH Stretching of amine), 3221 (NH Stretching of amine), 3047 (CH Stretching of aromatic ring), 2957 (CH Stretching of aliphatic), 1680 (C=O), 1574 (C=C), 1275 (C=S). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.03 (2H, s, 2NH), 10.87 (1H, s, NH), 7.14-7.49 (4H, m, aromatic), 3.57-3.63 (2H, m, CH_2), 1.62-1.66 (2H, m, CH_2), 1.15-1.48 (6H, m, $3CH_2$), 0.90 (3H, t, CH_3, J = 6.5 Hz,); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 14.4, 22.5, 26.6, 27.1, 31.3, 45.6, 114.9, 122.3, 135.1, 147.5, 163.1, 189.1; Anal Calcd for: $C_{15}H_{20}N_4OS$, C, 59.18; H, 6.62; N, 18.41; S, 10.53. Found: C, 59.46; H, 6.51; N, 18.53; S, 10.68.

N-(1H-benzimidazol-2-yl)-2-(pentylmino)-2-thioxoacetamide (4e)

Green crystals; Yield 72%; m.p. 198-200 °C; IR (KBr, cm^{-1}): 3256 (NH Stretching of amine), 3043 (CH Stretching of aromatic ring), 2958 (CH Stretching of aliphatic), 1680 (C=O), 1572 (C=C), 1275 (C=S). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.03 (2H, s, 2NH), 10.88 (1H, s, NH), 7.14-7.49 (4H, m, aromatic), 3.57-3.63 (2H, m, CH_2), 1.62-1.67 (2H, m, CH_2), 1.29-1.33 (2H, m, CH_2), 0.90 (3H, t, CH3, J = 6.7 Hz); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 14.3, 22.2, 26.8, 29.1, 45.5, 114.1, 122.3, 135.0, 147.5, 163.1, 189.1; Anal Calcd for: $C_{14}H_{18}N_4OS$, C, 57.91; H, 6.25; N, 19.29; S, 11.04. Found: C, 58.17; H, 6.33; N, 19.11; S, 11.33.

N-(1H-benzimidazol-2-yl)-2-(cyclopentylamino)-2-thioxoacetamide (4f)

Orange crystals; 60%; m.p. 180-182 °C; IR (KBr, cm^{-1}): 3256 (NH Stretching of amine), 3045 (CH Stretching of aromatic ring), 2957 (CH stretching of cyclopentyl), 1680 (C=O), 1564 (C=C), 1273 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.08 (2H, s, 2NH), 10.89 (1H, s, NH), 7.12-7.48 (4H, m, H-aromatic), 4.58-4.61 (1H, m, CH, cyclopentyl), 1.97-2.01 (2H, m, CH_2, cyclopentyl), 1.56-1.72 (6H, m, $3CH_2$, cyclopentyl); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 24.3, 31.3, 56.8, 114.0, 122.3, 134.7, 147.9, 164.6, 189.6; Anal Calcd for: $C_{14}H_{16}N_4OS$, C, 58.31; H, 5.59; N, 19.43; S, 11.12%. Found: C, 58.56; H, 5.51; N, 19.70; S, 11.33.

N-(1H-benzimidazol-2-yl)-2-(cyclohexylamino)-2-thioxoacetamide (4g)

Orange crystals; 57%; m.p. 219-221 °C; IR (KBr, cm^{-1}): 3385 (NH Stretching of amine), 3342 (NH Stretching of amine), 3059 (CH Stretching of aromatic ring), 2939 (CH stretching of cyclohexyl), 1678 (C=O), 1568 (C=C), 1273 (C=S). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): =), 12.01 (2H, s, 2NH), 10.68 (1H, s, NH), 7.13-7.47 (4H, m, aromatic), 4.14-4.22 (1H, m, CH), 1.14-1.92 (10H, m, $5CH_2$, cylohexyl); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 29.7, 30.2, 35.3, 59.2, 118.8, 127.1, 139.4, 152.8, 169.3, 193.8; Anal Calcd for: $C_{15}H_{18}N_4OS$, C, 59.58; H, 6.00; N, 18.53; S, 10.60. Found: C, 56.35; H, 5.89; N, 18.72; S, 10.75.

N-(1H-benzimidazol-2-yl)-2-(3-phenylpropylamino)-2-thioxoacetamide (4h)

Yellow crystals; Yield 51%; m.p. 173-175 °C; IR (KBr, cm^{-1}): 3315 (NH Stretching of amine), 3259 (NH Stretching of amine), 3026 (CH Stretching of aromatic ring), 2928 (CH stretching of aliphatic), 1678 (C=O), 1562 (C=C), 1273 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.03 (2H, s, 2NH), 10.91 (1H, s, NH), 7.14-7.49 (9H, m, aromatic), 3.58-3.65 (2H, m, CH_2), 2.63-2.68 (2H, m, CH_2), 1.91-2.01 (2H, m, CH_2); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm):

28.9, 33.0, 52.9, 114.2, 122.4, 126.3, 128.7, 128.8, 135.1, 141.8, 147.6, 163.2, 189.5; Anal Calcd for: $C_{18}H_{18}N_4OS$, C, 63.88; H, 5.36; N, 16.56; S, 9.47. Found: C, 64.20; H, 5.30; N, 16.29; S, 9.18.

N-(1H-benzimidazol-2-yl)-2-(benzylamino)-2-thioxoacetamide (4i)

Orange crystals; Yield 58%; m.p. 211-213 °C; IR (KBr, cm^{-1}): 3337 (NH Stretching of amine), 3259 (NH Stretching of amine), 3026 (CH Stretching of aromatic ring), 2931 (CH stretching of aliphatic), 1680 (C=O), 1562 (C=C Stretching of aromatic ring), 1273 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.10 (2H, s, 2NH); 11.21 (1H, s, NH), 7.14-7.48 (9H, m, aromatic), 4.85-4.87 (2H, d, CH_2, J = 5.9 Hz); Anal Calcd for: $C_{16}H_{14}N_4OS$, C, 61.92; H, 4.55; N, 18.05; S, 10.33. Found: C, 61.81; H, 4.61; N, 18.28; S, 10.02.

N-(1H-benzimidazol-2-yl)-2-(phenylamino)-2-thioxoacetamide (4j)

Orange crystals; Yield 53%; m.p. 221-223 °C; IR (KBr, cm^{-1}): 3342 (NH Stretching of amine), 3265 (NH Stretching of amine), 3072 (CH Stretching of aromatic ring), 1676 (C=O), 1572 (C=C Stretching of aromatic ring), 1271 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.31 (3H, br., 3NH), 7.16-7.96 (9H, m, H-aromatic),. ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 113.8, 120.9, 123.2, 123.7, 127.7, 129.3, 133.5, 138.4, 148.0, 163.8; Anal Calcd for: $C_{15}H_{12}N_4OS$, C, 60.79; H, 4.08; N, 18.91; S, 10.82. Found: C, 60.93; H, 4.19; N, 18.72; S, 10.51.

N-(1H-benzimidazol-2-yl)-2-(2-ethylanilino)-2-thioxoacetamide (4k)

Yellow crystals; Yield 60%; m.p. 228-230 °C; IR (KBr cm^{-1}): 3321 (NH Stretching of amine), 3259 (NH Stretching of amine), 3041 (CH Stretching of aromatic ring), 2931 (CH stretching of aliphatic), 1678 (C=O), 1568 (C=C Stretching of aromatic ring), 1273 (C=S). ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.28 (2H, s, 2NH), 12.06 (1H, s, NH), 7.08-7.50 (8H, m, H-aromatic), 2.50-2.70 (2H, m, CH_2), 1.13-1.24 (3H, t, CH_3, J = 7.4 Hz); Anal Calcd for: $C_{17}H_{16}N_4OS$, C, 62.94; H, 4.97; N, 17.27; S, 9.88. Found: C, 62.79; H, 5.23; N, 17.51; S, 9.56.

N-(1H-benzimidazol-2-yl)-2-(4-ethylanilino)-2-thioxoacetamide (4l)

Yellow crystals; Yield 55%; m.p. 226-228 °C; IR (KBr, cm^{-1}): 3248 (NH Stretching of amine), 3041 (CH Stretching of aromatic ring.), 2962 (CH stretching of aliphatic), 1676 (C=O), 1570 (C=C Stretching of aromatic ring), 1276 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.26 (3H, br., 3NH), 7.13-7.88 (8H, m, H-aromatic), 2.49-2.64 (2H, m, CH_2), 1.07-1.2 (3H, t, CH_3, J = 7.5 Hz); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 15.9, 28.3, 113.8, 122.5, 123.4, 128.3, 134.1, 136.8, 142.8, 148.8, 165.4, 189.6; Anal Calcd for: $C_{17}H_{16}N_4OS$, C, 62.94; H, 4.97; N, 17.27; S, 9.88. Found: C, 63.25; H, 5.18; N, 16.97; S, 9.59.

N-(1H-benzimidazol-2-yl)-2-(2-chloroanilino)-2-thioxoacetamide (4m)

Yellow crystals; Yield 50%; m.p. 183-185 °C; IR (KBr, cm^{-1}): 3331 (NH Stretching of amine), 3090 (CH Stretching of aromatic ring), 1680 (C=O), 1566 (C=C), 1273 (C=S) 742 (C-Cl); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.33 (3H, br., 3NH), 7.19-8.03 (8H, m, H-aromatic); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 113.4, 122.9, 127.5, 128.1, 129.0, 129,2 130.3, 132.7, 136.1, 149.9, 164.7, 191.8; Anal Calcd for: $C_{15}H_{11}ClN_4OS$, C, 54.53; H, 3.34; N, 16.96; S, 9.71. Found: C, 54.65; H, 3.40; N, 17.23; S, 9.53.

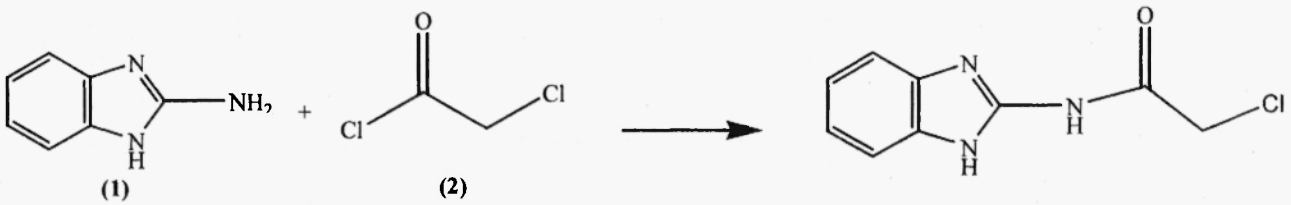
N-(1H-benzimidazol-2-yl)-2-(mesitylamino)-2-thioxoacetamide (4n)

Yellow crystals; Yield 50%; m.p. 227-229 °C; IR (KBr, cm^{-1}): 3209 (NH Stretching of amine), 3086 (CH Stretching of aromatic ring), 2986 (CH stretching of aliphatic), 1672 (C=O), 1570 (C=C), 1275 (C=S); ^{1}H-NMR (300 MHz, DMSO-d_6, δ / ppm): 12.24 (2H, br., 2NH); 11.92 (1H, br., NH), 7.16-7.50 (4H, m, aromatic), 6.95 (2H, s, aromatic), 2.28 (3H, s, CH_3), 2.14 (6H, s, $2CH_3$); ^{13}C-NMR (75 MHz, DMSO-d_6, δ / ppm): 17.9, 21.0, 113.8, 122.5, 129.0, 134.1, 134.6,

137.3, 148.4, 164.4, 178.3, 191.2; Anal Calcd for: $C_{18}H_{18}N_4OS$, C, 63.88; H, 5.36; N, 16.56; S, 9.48. Found: C, 63.61; H, 5.49; N, 16.39; S, 9.56.

RESULTS AND DISCUSSION

2-Aminobenzimidazole was reacted with chloroacetyl chloride to give *N*-(1*H*-benzimidazol-2-yl)-2-chloroacetamide **3**. Reaction of **3** with a mixture of corresponding amine and sulfur in DMF gave benzimidazolyl thioxoacetamides **4a-n** as shown in **Scheme 1**.



(4a)	R = $CH_3(CH_2)_2$ -	**(4h)**	R = $C_6H_5(CH_2)_3$-
(4b)	R = $CH_3(CH_2)_3$-	**(4i)**	R = (C_6H_5) CH_2-
(4c)	R = CH_3CH_2-CH(CH_3)	**(4j)**	R=C_6H_5-
(4d)	R = $CH_3(CH_2)_4$-	**(4k)**	R = 2-(C_2H_5) C_6H_4-
(4e)	R = $CH_3(CH_2)_5$-	**(4l)**	R = 4-(C_2H_5) C_6H_4-
(4f)	R = -$(CH_2)_4$ CH-	**(4m)**	R = 2-(Cl) C_6H_4-
(4g)	R = -$(CH_2)_5$ CH-	**(4n)**	R = 2,4,6-$(CH_3)_3$ C_6H_2-

Scheme 1

Compounds **4a-n** were synthesized using the variation of Zavarzin method (11). The reaction was proceeding smoothly under room temperature for 2-3 days depends on the used amines. Reaction with aromatic amines takes longer compared to the aliphatic amines. When aromatic amines with low basicity are used, triethylamine should be added to reaction mixture as a catalyst. Yield of products after recrstallization EtOH/H_2O were the order of 50-61%. The NMR spectra and elemental analysis data of all synthesized compounds are consistent with the expected structures. For example the ^{1}H-NMR spectrum of **4a** shows two broad singlets at 10.89 and 12.05 ppm, which attributed to the resonance of the three amine groups. Four aromatic protons related to two aromatic rings are appeared as a multiplet at 7.14-7.42 ppm. The signal at 0.91 ppm is due to resonance of the methyl group. The other aliphatic protons (2 CH_2 groups) resonate at 1.59-3.59 ppm.

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