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Recently we reported the reaction of 1,1-dimethyl-4-substituted semicarbazides with phosgene to occur with formation of several new heterocyclic ring systems. Here we report the reaction of several semicarbazides, thiosemicarbazides and related acyl substituted hydrazides with oxalyl chloride and malonyl chloride. These reactions occur with the formation of heterocyclic products which are oxadiazolinones, imidazolidinetriones, 2-thioxoimidazolidine-4,5-diones, and 2-thioxo-1,3-diazine-4,6-diones (2-thioxopyrimidine-4,6-diones).

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Introduction.

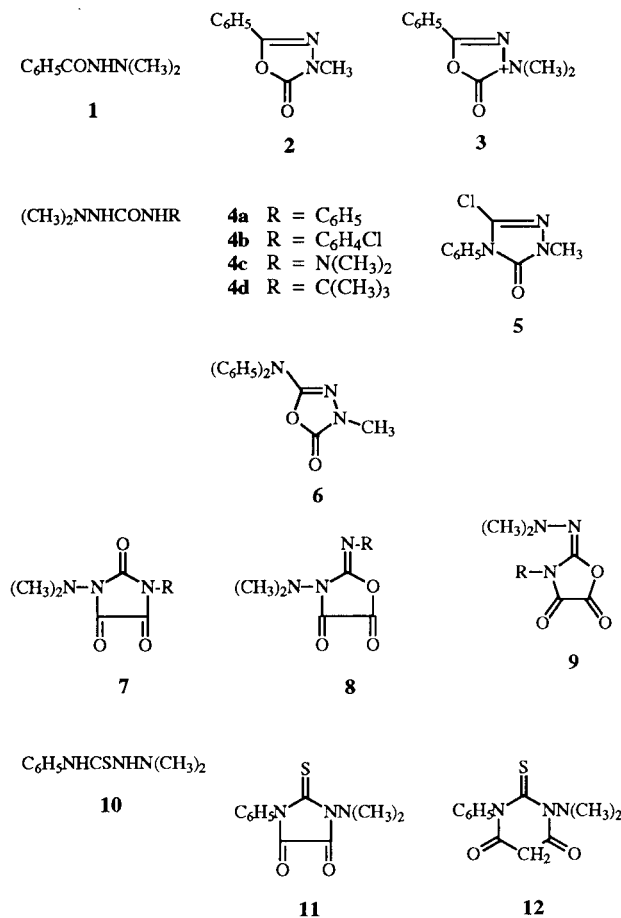
Recently we reported the reaction of 1,1-dimethyl-4-substituted semicarbazides with the diacylating agent, phosgene, to occur with the formation of several new heterocyclic ring systems [1]. Here we report the reaction of several acyl substituted hydrazides, semicarbazides, and thiosemicarbazides with the diacylating agents, oxalyl and malonyl chloride.

Reactions of hydrazines and hydrazides with acylating agents have been reviewed [2]. Several influences are observed. Acylation normally occurs on the more nucleophilic nitrogen. However, steric hindrance is important, and, if the more nucleophilic nitrogen is substituted with one or more large groups, acylation may occur on the less nucleophilic nitrogen. Dialkylation of one of the positions of hydrazine generally blocks acylation at that position, and 1,1-dimethylhydrazine, for example, undergoes mono- and di-acylation on the 2 position. In contrast, the diacylating agent, phosgene, reacts with 1-benzoyl-2,2-dimethylhydrazide (1) to form 4-methyl-2-phenyl- Δ^2 -1,3,4-oxadiazolin-5-one (2) [3]. The intermediate 3 was proposed for this reaction, and this intermediate would have to form by acylation on the dimethylated nitrogen of the hydrazide. A similar intermediate was proposed in the reaction of phosgene with 1,1-dimethyl-4-phenylsemicarbazide (4a) which gave 3-chloro-1-methyl-4-phenyl- Δ^2 -1,2,4-triazolin-5-one (5) [1].

Results and Discussion.

Treatment of 1,1-dimethyl-2-benzoylhydrazine with oxalyl chloride gave the corresponding hydrochloride of 1,1-dimethyl-2-benzoylhydrazine and the reported 4-methyl-2-phenyl- Δ^2 -1,3,4-oxadiazolin-5-one (2), which also forms in the reaction of 1 with phosgene [3] and by the lead tetraacetate oxidation of 1 [4,5]. Similarly, 1,1-dimethyl-4,4-diphenylsemicarbazide with oxalyl chloride

Scheme 1



afforded 2-diphenylamino-4-methyl- Δ^2 -1,3,4-oxadiazolin-5-one (6). With these two reactions both a dealkylation reaction [3,6] and a decarbonylation occur. A similar decarbonylation has been described in the preparation of acylisocyanates from amides and oxalyl chloride [7].

The reaction of **1** with oxalyl chloride was followed by nmr. After 24 hours, at room temperature a solution of **1** and oxalyl chloride with a 5 molar excess of oxalyl chloride in deuteriochloroform showed signals attributed to **2**, methyl chloride, and the hydrochloride of **1** [8]. The hydrochloride was isolated and characterized and found to react with oxalyl chloride to give **2**.

Reaction of 1,1-dimethyl-4-phenylsemicarbazide (**4a**), 1,1-dimethyl-4-*p*-chlorophenylsemicarbazide (**4b**), *N,N'*-bisdimethylamino-urea (**4c**) and 1,1-dimethyl-4-*tert*-butylsemicarbazide (**4d**) with excess oxalyl chloride occurred with the formation of products by loss of two moles of hydrogen chloride between the reactants. Decarbonylation and acylation on the dimethyl nitrogen did not occur in this reaction.

These reactions resemble the reactions reported for oxalyl chloride with ureas which were first reported a long time ago [9]. Three structures, **7**, **8**, and **9**, are possible for the product of this reaction. In papers appearing on this subject there has been little concern with the isomer question, and we felt it should be addressed. The preparation of the imidazolidinetrione (parabanic acid) has been reported by the reaction of oxalyl chloride or ethyl oxylate with urea [10]. The structure of imidazolidinetrione was proven by its formation by the oxidation of uric acid [11].

Stoffel [12] described the formation of substituted imidazolidinetriones from ureas with oxalyl chloride. In this same paper he suggested that thioureas react with oxalyl chloride to form an intermediate iminothiazolidinedione. The latter compounds rearrange to the 2-thioxoimidazolidine-4,5-diones on heating. Evidence presented for these structures was from the infrared spectra.

The ir and ^{13}C nmr data shown in Tables 1 and 2, respectively, are helpful in deciding which isomer is present.

nmr shows peaks at 154.6 for C-2, and 159.7 ppm for C-4 and C-5 [14]. Because these spectral data of the products were considered inconclusive, x-ray crystallography was used to prove that the structure of **7b** was correct for the product of **4b** with oxalyl chloride. The spectra of the products from **4** with oxalyl chloride are very similar, and the imidazolidinetrione structure is suggested for all of these.

Treatment of 1,1-dimethyl-4-phenyl thiosemicarbazide, **10**, with oxalyl chloride gave a yellow crystalline solid which was shown by mass spectrometry and combustion analysis to have formed by loss of two molecules of hydrogen chloride between the two reactants. The ir showed a strong broad absorption at 1774 cm^{-1} , while the ^{13}C nmr showed peaks at 153.5, 154.2 and 177.4 . For comparison 2-thioxoimidazolidine-4,5-dione was prepared using the published procedure [9b]. The ir showed a strong absorption at 1774 cm^{-1} , while the ^{13}C nmr

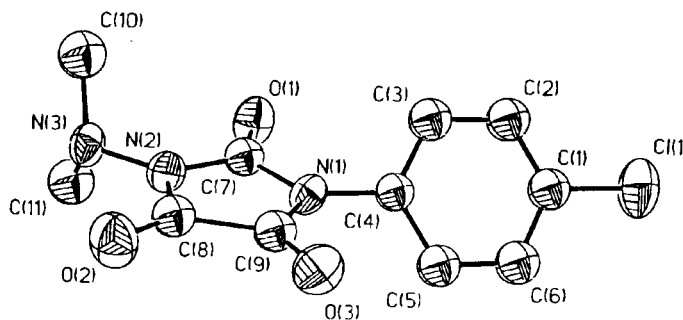


Figure 1. X-Ray Structural Analysis of **7b**.

Table 3
Atomic Coordinates ($\times 10^4$) and Equivalent Isotropic Displacement Coefficients for **7b** ($\text{\AA}^2 \times 10^3$)

Atom	x	y	z	U(eq)
Cl (1)	1525 (1)	5001 (2)	7291 (1)	78 (1)
N (1)	1164 (2)	3215 (5)	3144 (2)	40 (1)
N (2)	612 (2)	1691 (5)	1782 (2)	40 (1)
N (3)	125 (2)	361 (5)	1121 (2)	44 (1)
O (1)	80 (2)	466 (5)	3226 (2)	57 (1)
O (2)	1523 (2)	3536 (5)	704 (2)	61 (1)
O (3)	2268 (2)	5489 (4)	2506 (2)	56 (1)
C (1)	1459 (3)	4487 (7)	6067 (3)	51 (1)
C (2)	1588 (3)	2524 (7)	5763 (3)	54 (1)
C (3)	1520 (3)	2106 (8)	4781 (3)	51 (1)
C (4)	1297 (3)	3660 (6)	4146 (3)	40 (1)
C (5)	1174 (3)	5653 (7)	4458 (3)	49 (1)
C (6)	1268 (3)	6067 (8)	5424 (3)	54 (1)
C (7)	557 (3)	1642 (6)	2775 (3)	42 (1)
C (8)	1278 (3)	3136 (6)	1492 (3)	42 (1)
C (9)	1661 (3)	4158 (6)	2422 (3)	41 (1)
C (10)	466 (4)	-1745 (8)	1255 (4)	62 (1)
C (11)	-955 (4)	580 (9)	1125 (4)	63 (1)

*Equivalent isotropic U defined as one third of the trace of the trace of the orthogonalized U_{ij} tensor.

Table 1

IR Peaks in the C=X Region for **7a**, **7b**, **7c** and **7d**

	1827.5	1779.3	1744.6 (broad)
7a			
7b	1824.7	1777.4	1743.6 (broad)
7c	1816.9	1768.7	1741.7 (broad)
7d	1825.0	1772.0	1748.0 (broad)

Table 2

Chemical Shifts of C=X Carbon NMR Peaks for **7a**, **7b** and **7d**

	150.7	154.1	154.5
7a			
7b	150.5	153.3	154.0
7d	152.1	154.5	156.1

For comparison the ir of imidazolidinetrione shows peaks at 1842 and $1739\text{ (broad)}\text{ cm}^{-1}$ [13], while the ^{13}C

Table 4

Selected Bond Lengths (in Å) and Bond Angles (in degrees).

N(1)-C(4)	1.427(5)	N(1)-C(7)	1.393(5)
N(1)-C(9)	1.378(5)	N(2)-N(3)	1.403(4)
N(2)-C(7)	1.389(5)	N(2)-C(8)	1.373(5)
O(1)-C(7)	1.196(5)	O(2)-C(8)	1.189(5)
O(3)-C(9)	1.193(5)	C(8)-C(9)	1.523(6)
C(4)-N(1)-C(7)	123.6(3)	C(4)-N(1)-C(9)	125.4(3)
C(7)-N(1)-C(9)	110.8(3)	N(3)-N(2)-C(7)	126.3(3)
N(3)-N(2)-C(8)	121.5(3)	C(7)-N(2)-C(8)	112.0(3)
N(1)-C(7)-N(2)	106.9(3)	N(1)-C(7)-O(1)	126.7(4)
N(2)-C(7)-O(1)	126.3(4)	N(2)-C(8)-O(2)	129.3(4)
N(2)-C(8)-C(9)	104.4(3)	O(2)-C(8)-C(9)	126.2(4)
N(1)-C(9)-O(3)	127.3(4)	N(1)-C(9)-C(8)	105.6(3)
O(3)-C(9)-C(8)	127.1(4)		

showed two peaks at 162.1 and 184.2 ppm assigned to C=O and C=S, respectively. From these comparisons structure **11** is suggested.

When compounds **4a** and **4b** were treated with malonyl chloride, black intractable tars resulted. Treatment of **10** with malonyl chloride produced a crystalline solid which formed by loss of two molecules of hydrogen chloride between the two reactants. The ir showed two peaks in the carbonyl region at 1721 and 1708 cm^{-1} , while the ^{13}C nmr showed peaks at 162.5, 163.0 and 200.0. Structure **12**, 1-dimethylamino-3-phenyl-2-thioxo-4,6(1*H*,5*H*)-pyrimidinedione, is indicated from these data.

EXPERIMENTAL

^1H and ^{13}C nmr spectra were obtained, in the indicated solvent, from either an IBM NR/300 FTNMR or a JEOL FX90Q spectrometer; reported chemical shifts are in ppm (δ) relative to either chloroform (δ 7.26) or TMS (δ 0.00). Infrared spectra were recorded on a Digilab Qualimatic FTIR instrument using sodium chloride plates. The ms measurements were made with a VG 7070HS mass spectrometer. Melting points were determined by using a Thomas-Hoover apparatus and were corrected. Elemental analyses were carried out by Desert Analytics, Tucson, AZ.

Reagents were purchased from Aldrich Chemical Co. Inc., Milwaukee, WI. Solvents were glass distilled and were obtained from either Burdick and Jackson Laboratories, Inc., Muskegon, MI, or from EM Science, Cherry Hill, NJ.

Crystal Structure Analysis of **7b**.

A single crystal was mounted on a Nicolet R3m/E diffractometer equipped with a graphite monochromator. Lattice constants of this monoclinic crystal were determined to be $a = 13.426$ (5) Å, $b = 6.524$ (2) Å, $c = 13.919$ (3) Å, $\beta = 92.75$ (2)° with $\text{MoK}\alpha$ radiation (0.710688 Å). Data collection at ambient temperature yielded 2114 unique reflections. Structure solution *via* direct methods and subsequent different Fourier synthesis yielded the positions of the C, N, O, and H atoms of the molecule. Least-squares refinement with hydrogen atoms included at calculated positions gave a final R value of 0.0682 for data with $|F| \geq 3\sigma(F)$.

2-Phenyl-4-methyl- Δ^2 -1,3,4-oxadiazolin-5-one (**2**) and 1,1-Dimethyl-2-benzoylhydrazide Hydrochloride.

To 1,1-dimethyl-2-benzoylhydrazide [14] (0.0164 g, 0.1 mmole) dissolved in approximately 1 ml of deuteriochloroform in an nmr tube was added oxalyl chloride (0.041 ml, 0.5 mmole). After 1 hour at 25° a white precipitate began to form. On warming to 40° this precipitate dissolved. The nmr spectrum taken 24 hours later at 40° revealed signals for **2**, methyl chloride and 1,1-dimethyl-2-benzoylhydrazide hydrochloride in a 67:67:33 ratio. The solid product obtained by evaporating this chloroform solution weighed 0.0118 g, 62%. The mixture was separated by column chromatography on silica gel and **2**, identical with the known by ir, nmr, R_f and mp, eluted with ethyl acetate. A second solid product eluted more slowly from silica gel with 4:1 benzene methanol solvent, mp 180-182°; ir (Nujol mull): ν 1686 (C=O) cm^{-1} ; ^1H nmr (DMSO- d_6): δ 3.44 (6H, s), 7.24-7.94 (5H, m), 12.3 (1H, s); ^{13}C nmr: δ 47.1, 128.3, 128.5, 129.4, 133.2, and 165.2; ms: 165 (M^+ , 100). When this product was treated with oxalyl chloride, it gradually gave a mixture of **2** and methyl chloride.

Anal. Calcd. for $\text{C}_9\text{H}_{13}\text{N}_2\text{OCl}$: C, 53.86; H, 6.53; N, 13.96; Cl, 17.66. Found: C, 53.75; H, 6.53; N, 13.90; Cl, 17.63.

1,1-Dimethyl-4,4-diphenylsemicarbazide.

This semicarbazide was prepared by adding a 1 molar chloroform solution of diphenylcarbamoyl chloride to a rapidly stirred, ice-cooled 1 molar chloroform solution of 1,1-dimethylhydrazine. During the addition a precipitate appeared. After addition was complete, the slurry which resulted was stirred at room temperature for 12 hours, washed with successive portions of saturated sodium bicarbonate solution until the washings remained basic and dried over magnesium sulfate. Removal of the drying agent and the solvent in the usual way afforded (96%) white needles (light sensitive) which were recrystallized from a mixture of dichloromethane (soluble) and hexane (insoluble), mp 130-131.5°; ir (potassium bromide): ν 1670 (C=O) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.59 (s, 6H), 5.48 (broad s, 1H), 7.26 (s, 10H); ms: m/z (relative intensity) 255 (M^+ , 22), 212 (27), 211 (24), 196 (28), 169 (37), 168 (37), 166 (32), 93 (33), 77 (36), 59 (100), 43 (49), 42 (33).

Anal. Calcd. for $\text{C}_{15}\text{H}_{17}\text{N}_3\text{O}$: C, 70.56; H, 6.71; N, 16.46. Found: C, 70.22; H, 6.58; N, 16.46.

2-Diphenylamino-4-methyl- Δ^2 -1,3,4-oxadiazolin-5-one (**6**). Phosgene Method.

1,1-Dimethyl-4,4-diphenylsemicarbazide was treated with excess phosgene in the same manner as described earlier [1]. Compound **6** was recrystallized from ethyl acetate (73%), mp 158.5-159.5°; ir (potassium bromide): ν 1793 (C=O) and 1625 (C=N) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 3.32 (s, 3H), 7.16-7.44 (m, 10H); ms: m/z (relative intensity) 267 (M^+ , 100), 222 (2.2), 196 (18.5), 180 (24.7), 168 (34.3), 167 (22.3), 77 (65.8), 51 (33.7).

Anal. Calcd. for $\text{C}_{15}\text{H}_{13}\text{N}_3\text{O}_2$: C, 67.41; H, 4.90; N, 15.72. Found: C, 67.53; H, 5.01; N, 15.73.

2-Diphenylamino-4-methyl- Δ^2 -1,3,4-oxadiazolin-5-one (**4**). From Oxalyl Chloride.

A 300-ml flask was charged with 1,1-dimethyl-4,4-diphenylsemicarbazide (2.321 g, 9.10 mmoles) and dichloromethane (125 ml). The flask was cooled to 0°

(ice/water) under nitrogen, and oxalyl chloride (3.97 ml, 45.5 mmoles) was added *via* a syringe. An immediate yellow color formed which persisted. The solution was allowed to warm to room temperature and was stirred for 24 hours. The contents were poured into 200 ml of water. The layers were separated and the aqueous portion extracted with dichloromethane (2 x 70 ml). The combined organic layers were dried (magnesium sulfate), and the solvent removed under reduced pressure. The resulting white solid (1.638 g, 6.14 mmoles, 67%) was found to be identical in all respects (ir, nmr, ms undepressed mixed mp) to the product formed with excess phosgene.

1,1-Dimethylamino-3-substituted Imidazolidinetriones. General Method, 7a-7c.

The semicarbazides were prepared as described in the literature: **4a** [1], **4b** [15], **4c** [1], **4d** [16]. A solution of 10 mmoles of **4** in 50 ml of methylene chloride was chilled in an ice-water bath under a nitrogen atmosphere. Oxalyl chloride, 50 mmoles, was added by syringe, and the solution was kept cold for 30 minutes. The solution was then stirred for two days after which time the solvent was removed.

1,1-Dimethylamino-3-phenylimidazolidinetrione (7a).

Analysis (tlc silica gel diethyl ether) showed two spots ($R_f = 0.00, 0.50$) neither of which corresponded to **4a** ($R_f = 0.22$). The product, $R_f = 0.50$, was isolated *via* column chromatography (1.90 g, 8.15 mmoles) 82% and recrystallized from absolute ethanol, mp 129.5-130°; ir (melt): ν 1826, 1773, 1749 (br), 1653 (sh) (C=O) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.81 (s, 6H), 7.25 (s, 5H); ^{13}C nmr (deuteriochloroform): δ 44.6, 125.6, 128.9, 129.2, 129.4, 150.7, 154.1, 154.5; ms: m/z (relative intensity) 233 (M^+ , 16.2), 190 (2.4), 119 (5.1), 91 (7.0), 86 (100), 70 (7.3), 64 (6.2), 58 (44.6), 43 (73.0).

Anal. Calcd. for $\text{C}_{11}\text{H}_{11}\text{N}_3\text{O}_3$: C, 56.65; H, 4.75; N, 18.02. Found: C, 56.74; H, 4.79; N, 18.03.

1,1-Dimethylamino-4-*p*-chlorophenylimidazolidinetrione (7b).

The product (2.1123 g, 79%) was recrystallized from toluene, mp 147-148°; ir (Nujol): ν 1740 (C=O); ^1H nmr (deuteriochloroform): δ 2.98 (s, 6H), 7.23-7.46 (m, 4H); ^{13}C nmr (deuteriochloroform): δ 44.8 (NCH₃), 127, 128, 129.5, 135 (aromatic), 150.5, 153.3, 154 (C=O); ms: m/z (relative intensity) 269 (3.00) ($\text{M}+2^+$), 267 (9.07) (M^+), 155 (12.17), 154 (3.07), 153 (39.59), 127 (5.52), 125 (14.95), 90 (15.26), 89 (3.81), 87 (3.12), 86 (100), 85 (2.47), 76 (3.07), 75 (5.31), 70 (7.37), 64 (4.08) 63 (11.16), 58 (36.43). To grow a crystal suitable for X-ray analysis the sample was dissolved in acetone and placed in an atmosphere saturated with pentane.

Anal. Calcd. for $\text{C}_{11}\text{H}_{10}\text{N}_3\text{O}_3\text{Cl}$: C, 49.35; H, 3.76; N, 15.69. Found: C, 49.55; H, 3.63; N, 15.68.

1,1-Dimethylamino-3-*tert*-butylimidazolidinetrione (7c).

This product was purified by recrystallization from ethanol-pentane mixture giving beautiful, long, needle-shaped crystals, 1.6241 g (76%), mp 110-111°; ir (Nujol): ν 1816.9, 1768.7, 1741.7 (C=O) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.98 (s, 6H), 1.59 (s, 9H); ^{13}C nmr (deuteriochloroform): δ 28.7, 44.6, 60.3, 152, 154, 156; ms: m/z (relative intensity) 213 (7.8), 157 (26.7), 86 (100). The tlc on silica gel using ethyl acetate showed one spot with R_f of 0.60.

Anal. Calcd. for $\text{C}_9\text{H}_{15}\text{N}_3\text{O}_3$: C, 50.69; H, 7.04; N, 19.71. Found: C, 50.70; H, 7.04; N, 19.90.

1,3-Bis(dimethylamino)imidazolidinetrione (7d).

The general procedure described above was used with the following changes. The proportion of reactants was changed to a ratio of 2.42 mmoles of *N,N*-bisdimethylaminourea, **4c** [16], and 2.42 mmoles of oxalyl chloride. It was necessary to add 4.84 mmoles of pyridine as well [17], and to increase the amount of methylene chloride to 150 ml. The reaction time was decreased to 5 hours. The product was separated from the pyridinium hydrochloride by column chromatography (silica, ethyl acetate). Removal of the solvent left 0.320 g (67%) of a white solid which was recrystallized from carbon tetrachloride, mp 118-119°; ir (melt): ν 1825, 1772, 1748 (br) (C=O) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.96 (s); ms: m/z (relative intensity) 200 (M^+ , 46.7), 158 (2.3), 149 (2.4), 129 (2.8), 86 (100), 70 (10.7), 58 (48.2), 43 (78.1).

Anal. Calcd. for $\text{C}_7\text{H}_{12}\text{N}_4\text{O}_3$: C, 42.00; H, 6.04; N, 27.99. Found: C, 41.73; H, 5.98; N, 27.76.

1,1-Dimethylamino-3-phenylthioximiclazolidine-4,5-dione (11).

The 1,1-dimethyl-4-phenyl thiosemicarbazide, **10**, was prepared as described in the literature [18]. A solution of 1,1-dimethyl-4-phenylthio semicarbazide (2.50 g, 12.80 mmoles) in 100 ml of methylene chloride was chilled in an ice-water bath under a nitrogen atmosphere. Oxalyl chloride (5.58 ml, 64.0 mmoles) was added by a syringe, and the solution was kept cold for 30 minutes. The solution was then stirred for two days after which time the solvent was removed leaving a yellow precipitate. The product was purified by column chromatography (silica gel, ethyl acetate) and recrystallized from carbon tetrachloride and obtained as yellow crystals, mp 57-58°; ir (Nujol): ν 1774 cm^{-1} (CO); ^1H nmr (deuteriochloroform): δ 3.05 (s, 6H), 7.23-7.52 (m, 5H) ppm; ^{13}C nmr (deuteriochloroform): δ 44.1 (N-CH₃), 128.0, 129.4, 129.9, 131.5 (aromatics), 153.5 (C=O), 154.2 (C=O), 177.4 (C=S) ppm; ms: m/z (relative intensity) 249 (M^+ , 18.85), 208 (100), 193 (6.76), 188 (1.01), 178 (16.21).

Anal. Calcd. for $\text{C}_{11}\text{H}_{11}\text{N}_3\text{O}_2\text{S}$: C, 52.99; H, 4.44; N, 16.86. Found: C, 53.17; H, 4.38; N, 16.97.

1,1-Dimethylamino-3-phenyl-2-thioxo-4,6(1H,5H)-pyrimidine-dione (12).

A 250 ml round-bottomed flask was equipped with a stirring bar and charged with 1,1-dimethyl-4-phenyl thiosemicarbazide, **10**, [18] (5.0 g, 25.60 mmoles) and dichloromethane (150 ml). The contents were cooled to 0° (ice/water) under nitrogen. Then malonyl chloride (2.5 g, 25.60 mmoles) was added *via* a syringe. After 30 minutes the cold bath was removed and the solution was stirred for 72 hours. The solvent was removed, and the product separated by column chromatography (silica gel, methylene chloride). Removal of the solvent left 3.25 g (48%) of a dark brown solid which was recrystallized from hexane and chloroform mixture and formed needle-shaped crystals, mp 162-164°; ir (Nujol): ν CO 1721, 1708 (C=O) cm^{-1} ; ^1H nmr (deuteriochloroform): δ 2.96 (s, 6H), 3.92 (s, 2H), 7.09-7.52 (m, 5H); ^{13}C nmr (deuteriochloroform): δ 42.0 (N-CH₃), 42.8 (-CH₂-), 128.5 (aromatic), 128.8 (aromatic), 129.6 (aromatic), 162.5 (C=O), 163.0 (C=O), 200 (C=S); ms: m/z (relative intensity) 263 (M^+ , 5.63), 221 (58.90), 192 (3.07), 177 (3.71), 162 (2.81), 145 (17.92), 119 (51.85), 101 (14.56), 91 (44.43), 86 (65.04), 77 (100), 69 (48.78), 65 (33.93), 58 (32.13).

Anal. Calcd. for $\text{C}_{12}\text{H}_{13}\text{N}_3\text{O}_2\text{S}$: C, 54.73; H, 4.97; N, 15.96. Found: C, 54.70; H, 4.90; N, 15.96.

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