Reaction of Chloropyridazines with *N,N*-Dimethylformamide

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Received February 14, 2000

Chloropyridazine derivatives **1**, **3**, **5**, **7** and **10a-c** were reacted with *N,N*-dimethylformamide under reflux condition to give the corresponding *N,N*-dimethylaminopyridazines **2**, **4**, **6**, **8**, **9** and **11a-c** regioselectively.

J. Heterocyclic Chem., **37**, 1591 (2000).

In connection with our research program for the synthetic application of novel heterocycles containing pyridazine, we required various *N,N*-dimethylaminopyridazines.

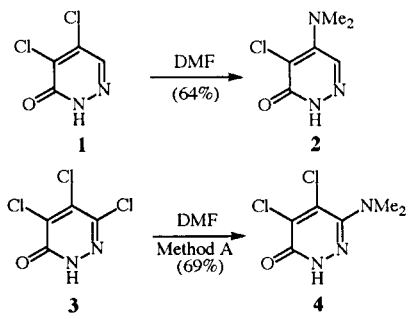
Some methods of *N,N*-dimethylamination for an aromatic or a heteoaryl halide have been reported by Kuroda [1], Lawesson [2], Ohta [3] and Gupton groups [4].

On the other hand, *N,N*-dimethylaminations of reactive aryl or benzyl halide with *N,N*-dimethylformamide, either in the presence or absence of catalysts such as copper salts, have been reported [5].

In a previous paper [6], we reported the reaction of 4,5-dichloro-6-nitro-2*H*-pyridazin-3-one with dimethylchloromethyleneammonium chloride in *N,N*-dimethylformamide to give the corresponding 6-(*N,N*-dimethylamino)derivative. Therefore, we attempted the investigation of the *N,N*-dimethylamination of chloropyridazines with *N,N*-dimethylformamide.

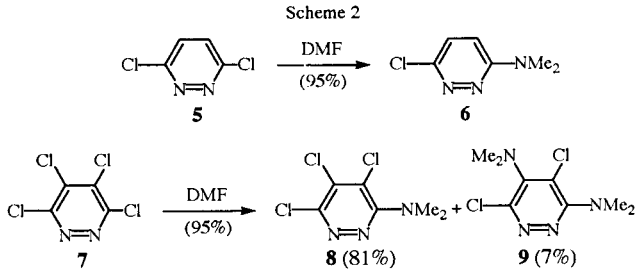
In this paper, we would like to report the results of the title reaction. The reaction of 4,5-dichloro-2*H*-pyridazin-3-one (**1**) or 4,5,6-trichloro-2*H*-pyridazin-3-one (**3**) with refluxing *N,N*-dimethylformamide afforded the *N,N*-dimethylaminopyridazines **2** (64%) or **4** (69%) (Scheme 1), regioselectively. 3,6-Dichloropyridazine (**5**) also was treated with refluxing *N,N*-dimethylformamide to give only **6** in excellent yield, whereas 3,4,5,6-tetrachloropyridazine (**7**) was reacted with refluxing *N,N*-dimethylformamide to yield compounds **8** (81%) and **9** (7%) (Scheme 2).

Scheme 1



1 **2** **3** **4**

Scheme 2



5 **6** **7** **8** (81%) **9** (7%)

Table 1

Melting Points and Infrared Spectral Data of **2**, **4**, **6**, **8**, **9**, **11a-c**, **12**, **13** and **14**

Compound No	Mp (°C) (lit. mp)	IR (Potassium bromide) (cm^{-1})
2	191-193 (200-201)[16]	3170, 3100, 2950, 1670, 1630, 1530, 1450, 1380, 1300, 1260, 1240, 1180, 1160, 900, 880, 780, 750, 640, 600
4	183 (193-194)[6]	3220, 3150, 3050, 2980, 2920, 1680, 1580, 1430, 1280, 1160, 1000, 940, 880, 700, 640
6	90-91 (104-106)[17]	3080, 2950, 1610, 1540, 1510, 1438, 1410, 1225, 1180, 1080, 1020, 960, 840, 790, 620
8	80-81	2950, 2910, 1540, 1460, 1438, 1395, 1310, 1260, 1195, 1070, 980, 850, 795, 615
9	51-52	2950, 2915, 2830, 1555, 1520, 1460, 1380, 1260, 1200, 1040, 860, 800
11a	101-103	3100, 3050, 3000, 2875, 1620, 1570, 1520, 1450, 1430, 1312, 1260, 1220, 1120, 1050, 980, 900, 860, 830, 780, 700
11b	128-130	3100, 2970, 2910, 2250, 1630, 1560, 1520, 1450, 1330, 1308, 1250, 1190, 1130, 1080, 1040, 980, 900, 860
11c	138-141	3100, 2990, 2940, 1630, 1610, 1580, 1530, 1510, 1460, 1430, 1385, 1295, 1240, 1190, 1130, 1080, 1040, 980, 900, 850, 700, 680
12	73-74 (74-75)[7, 18]	3110, 2960, 2840, 1660, 1610, 1520, 1465, 1405, 1360, 1325, 1210, 1070, 880
13	110-111	3046, 2919, 1628, 1599, 1530, 1439, 1413, 1359, 1338, 1292, 826
14	231-233	3050, 2950, 1638, 1420, 1370, 1180

On the other hand, we attempted the dimethylamination of the 3-chloro-6-phenoxy derivatives **10a-c** with *N,N*-dimethylformamide. Reaction of **10a** with only refluxing *N,N*-dimethylformamide did not afford the corresponding *N,N*-dimethylamino derivatives.

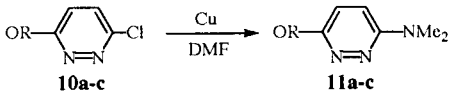
Table 2
^{1}H NMR Spectral Data of **2**, **4**, **6**, **8**, **9**, **11a-c**, **12**, **13** and **14**

Compound No	Solvent [a]	^{1}H NMR (δ, ppm) [b] $N(CH_3)_2$ (s)	Others
2	D	3.31	7.81 (s, 1H), 12.7 (s, bs, NH)
4	D	3.16	NH (No detection)
6	C	3.08	6.70 (d, 1H, J = 9.5), 7.09 (d, 1H, J = 9.6)
8	C	3.09	—
9	C	2.93 2.98	—
11a	C	3.13	3.81 (s, 3H), 6.90 (d, 2H, J = 9.0), 6.96 (d, 1H, J = 8.9), 6.99 (d, 1H, J = 8.9), 7.81 (d, 2H, J = 8.9)
11b	C	3.16	6.91 (d, 1H, J = 9.6), 6.98 (d, 1H, J = 9.6), 7.20 (d, 2H, J = 8.7), 7.57 (d, 2H, J = 8.7)
11c	C	3.17	7.01 (d, 1H, J = 9.7), 7.08 (d, 1H, J = 9.6), 7.31 (d, 2H, J = 9.2), 8.24 (d, 2H, J = 9.2)
12	C	3.10	3.73 (s, 3H), 7.55 (s, 1H)
13 [7]	C	3.01	3.70 (s, 3H), 5.73 (d, 1H, J = 2.9), 7.57 (d, 1H, J = 2.9)
14	D	3.18 3.22	6.02 (d, 1H, J = 1.8), 8.37 (d, 1H, J = 1.8)

[a] D = Dimethyl-d_6 sulfoxide, C = Deuteriochloroform. [b] Abbreviations used: s = singlet, bs = broad singlet, d = doublet. J = Hz unit. The proton signal of NH group was exchangeable with deuterium oxide.

N,N-Dimethylamination of **10a-c**, however, with refluxing *N,N*-dimethylformamide in the presence of copper powder as a catalyst gave 3-(*N,N*-dimethylamino) derivatives **11a-c** in low yield instead of the corresponding Ullman products such as bipyridazines (Scheme 3).

Scheme 3



10a-c **11a-c**

11a: R = *p*-Methoxyphenyl (22%)
11b: R = *p*-Cyanophenyl (14%)
11c: R = *p*-Nitrophenyl (36%)

Table 3
^{13}C NMR Spectral Data of **2**, **4**, **6**, **8**, **9**, **11a-c**, **12**, **13** and **14**

Compound No	Solvent [a]	^{13}C NMR (δ, ppm)
2	D	42.1, 110.0, 131.5, 148.6, 159.1
4	D	42.4, 116.0, 138.4, 146.3, 158.2
6	C	38.6, 114.4, 128.7, 146.0, 159.2
8	C	43.3, 129.6, 147.6, 152.6, 155.8
9	C	42.1, 43.0, 122.9, 147.3, 147.7, 162.5
11a	C	38.5, 55.6, 114.6, 116.5, 119.3, 121.8, 148.0, 156.3, 157.9, 159.9
11b	C	38.5, 99.0, 107.3, 116.5, 118.7, 120.1, 120.5, 133.8, 158.0, 158.5
11c	C	40.9, 118.9, 122.2, 122.6, 127.9, 146.0, 160.4, 160.9, 162.6
12	C	40.6, 42.2, 113.0, 129.9, 148.3, 159.2
13	C	39.5, 39.7, 99.0, 128.1, 150.1, 162.2
14	D	39.9, 40.1, 89.5, 133.1, 148.5, 152.6

[a] D = Dimethyl-d_6 sulfoxide, C = Deuteriochloroform.

We attempted to determine the position of the *N,N*-dimethylamino group in the structure **2** using the coupling constants for C4-H and C6-H of the proton magnetic resonance of 5-amino-2*H*-pyridazin-3-one. Because of the low solubility of compound **2**, however, we selected the corresponding 2-methyl isomers **12** as the starting material for the dehalogenation. According to Kweon's method [6], the reaction of **2** with methyl iodide in the presence of potassium carbonate in acetone yielded compound **12** in 67% yield. Treatment of **12** with Pd/C and hydrogen in the presence of aqueous sodium hydroxide (10%) in ethanol gave 5-(*N,N*-dimethylamino)-2-methyl-2*H*-pyridazin-3-one (**13**) in 37% yield. The ^{1}H nmr spectrum of **13** showed the signals of two aromatic protons at δ 5.73 and δ 7.57 ppm as doublets and the proton signals of the three methyl groups, respectively. The chemical shift values, the spectral pattern and the coupling constant ($J_{4,6}$) of two aromatic protons for **13** were identical with the literature values [7,8,9]. Therefore, the position of the *N,N*-dimethylamino group for **2** may be regarded as a 5-substituted isomer as in structure **2**.

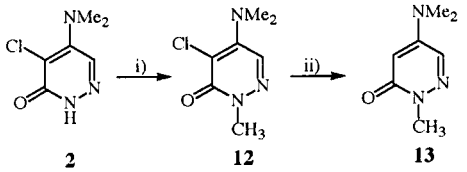
On the other hand, reaction of **8** with glacial acetic acid under refluxing conditions gave **4** in 30% yield (Method B in Scheme 5). This compound **4** was also identical with the products that were prepared by the Method A and Kweon's Method [6]. Therefore, the position of the *N,N*-dimethylamino group for **8** may be regarded as at 3-position.

In order to establish the structure of **9**, compound **9** was dehalogenated with Pd/C under hydrogen atmosphere to give 3,5-di-(*N,N*-dimethylamino)pyridazine (**14**) in 87% yield (Scheme 6). The ^{1}H nmr spectrum showed the signals of the two aromatic protons at δ 6.02 and δ 8.37 ppm as doublets, respectively. The coupling constant value for $J_{4,6}$ of the two aromatic protons in the structure **14** is 1.8 Hz. In accordance with the literatures [10,11,12], the coupling constant between C4-H and C5-H for 3,6-disubstituted-pyridazines is about 8 - 10 Hz.

On the other hand, Katz, *et al.* [8] and Kweon, *et al.* [9] reported that the coupling constant between C4-H and C6-H is smaller than that between C5-H and C6-H ($J_{4,6}$ = about 2 Hz, $J_{5,6}$ = about 5 Hz) for 2-substituted-5-(or 4-)-monosubstituted-pyridazin-3-one. Bryant *et al.* [13] and Kweon *et al.* [14] reported that the coupling constant between C4-H and C6-H for 3,5-disubstituted-pyridazines is about 2.3 - 2.8 Hz. Therefore, the position of both *N,N*-dimethylamino groups may be regarded as at the 3-position and the 5-position as in the structure **9**.

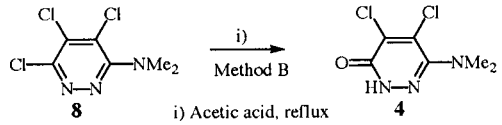
According to the literature [15], the *N,N*-dimethylamination of chloropyridazines may be regarded that it occurs *via* a decomposition of *N,N*-dimethylformamide. *N,N*-Dimethylformamide decomposes slowly at the reflux temperature into carbon monooxide and *N,N*-dimethylamine.

Scheme 4



i) MeI, K_2CO_3, Acetone ii) H_2, Pd/C, NaOH (10%), Ethanol.

Scheme 5



i) Acetic acid, reflux

Scheme 6

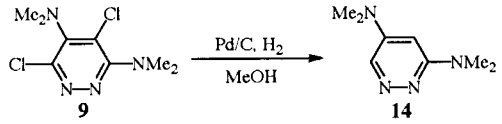


Table 4
Elemental Analytical Data of **2**, **4**, **6**, **8**, **9**, **11a-c**, **12**, **13** and **14**

Compound No.	Molecular Formula	Analysis (%) Calcd./Found		
		C	H	N
2	$C_6H_8N_3OCl$	41.51	4.64	24.20
		41.31	4.38	24.13
4	$C_6H_7N_3OCl_2$	34.64	3.39	20.20
		34.65	3.27	20.20
6	$C_6H_8N_3Cl$	45.73	5.12	26.66
		45.90	5.35	26.45
8	$C_6H_6N_3Cl_3$	31.82	2.67	18.55
		31.62	2.55	18.45
9	$C_8H_{12}N_4C_{12}$	40.87	5.14	23.83
		41.12	5.37	23.68
11a	$C_{13}H_{15}N_3O_2$	63.66	6.16	17.13
		63.88	5.91	17.01
11b	$C_{13}H_{12}N_4O_2$	60.93	4.72	21.86
		60.68	4.82	21.98
11c	$C_{12}H_{12}N_4O_3$	55.38	4.65	21.53
		55.47	4.87	21.58
12	$C_7H_{10}N_3OCl$	44.81	5.37	22.40
		44.78	5.40	22.62
13	$C_7H_{11}N_3O$	54.89	7.24	27.43
		54.95	7.14	27.63
14	$C_8H_{14}N_4$	57.81	8.49	33.71
		57.81	8.32	33.59

Because of the reaction of chloropyridazines with *N,N*-dimethylamine is generated by the decomposition of *N,N*-dimethylformamide, the *N,N*-dimethylamination of chloropyridazines under these conditions needs both reflux temperature and long reaction times. According to our observation, the quality of *N,N*-dimethylformamide did not have any effect on either the yield or the reaction time.

In conclusion, *N,N*-dimethylformamide is useful for the conversion of chloropyridazines to the corresponding *N,N*-dimethylaminopyridazines. Our method is also simple, efficient and regioselective. The use of *N,N*-dimethylformamide as a solvent, however, in nucleophilic displacements involving halopyridazines is not recommended.

Further work including the theoretical study on the regioselectivity, the application and other chemical transformations of the products are under way in our laboratory.

EXPERIMENTAL

Melting points were determined with a Thomas-Hoover capillary apparatus and are uncorrected. Magnetic resonance spectra were obtained on a Bruker FTNMR-DRX 500 spectrometer with chemical shift values reported in δ units (part per million) relative to an internal standard (tetramethylsilane). Infrared spectral data were obtained on a Hitachi 270-50 spectrophotometer. Elemental analyses were performed with a Perkin Elmer 240C. Open-bed column chromatography was carried out silica gel 60 (70-230 mesh, Merck) using gravity flow. The column was packed as slurries with the elution solvent. The substrates **10a-c** [8] were prepared by reported methods. *N,N*-Dimethylformamide (above 99%) was also purified by distillation in the presence of molecular sieves (4 Å).

4-Chloro-5-(dimethylamino)-2*H*-pyridazin-3-one (**2**).

A solution of **1** (500 mg, 3.03 mmoles) in *N,N*-dimethylformamide (10 ml) was refluxed for 48 hours. After cooling to room temperature, the mixture was poured into ice water (100 ml) with stirring. The product was extracted with chloroform (25 ml x 2). The chloroform solution was then dried over anhydrous magnesium sulfate. The organic solution was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing the product were combined and then evaporated under reduced pressure. Cold water (100 ml) was added to the residue with stirring. The resulting precipitates were filtered and dried in air to give compound **2** (237.7 mg, 64%) as colorless needles.

4,5-Dichloro-6-(dimethylamino)-2*H*-pyridazin-3-one (**4**).

Method A.

A solution of compound **3** (500 mg, 2.39 mmoles) in *N,N*-dimethylformamide (10 ml) was refluxed for 48 hours. After cooling to room temperature, the mixture was poured into ice water (150 ml) with stirring. The product was extracted with chloroform (25 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solvent was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing the product were combined and then evaporated under reduced pressure. Cold water (100 ml) was added to the residue with stirring. The resulting precipitates were filtered and dried in air to give compound **4** (343 mg, 69%) as colorless needles.

Method B.

A solution of **8** (200 mg, 0.88 mmole) and glacial acetic acid (5 ml) was refluxed for 1 hour. The acetic acid was removed under reduced pressure. The residue was triturated in water,

filtered and recrystallized from ethanol to give **4** (30 mg, 33%) as colorless needles.

6-Chloro-3-(dimethylamino)pyridazine (**6**).

A solution of **5** (500 mg, 3.38 mmoles) in *N,N*-dimethylformamide (10 ml) was refluxed for 10 hours. After cooling to room temperature, the mixture was poured into ice water (100 ml) with stirring. The product was extracted with chloroform (20 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solution was evaporated under reduced pressure. The resulting residue was purified by column chromatography on silica gel (elution solvent: chloroform) to give **6** (506 mg, 95%) as colorless needles.

3-(Dimethylamino)-4,5,6-trichloropyridazine (**8**) and 3,5-Dichloro-4,6-di-(dimethylamino)pyridazine (**9**).

A solution of compound **7** (1.9 g, 8.72 mmoles) in *N,N*-dimethylformamide (15 ml) was refluxed for 3 hours. After cooling to room temperature, the mixture was poured into ice water (100 ml) with stirring. The product was extracted with chloroform (20 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solution was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing **8** (R_f = 0.8, chloroform) were combined and evaporated under reduced pressure to give **8** (1.6 g, 81%). Fractions containing **9** (R_f = 0.7, chloroform) were combined and evaporated under reduced pressure to afford **9** (126 mg, 7%) as colorless needles.

3-(Dimethylamino)-6-(4-methoxyphenoxy)pyridazine (**11a**).

A mixture of **10a** (500 mg, 1.98 mmoles) [10] in *N,N*-dimethylformamide (10 ml) in the presence of Cu powder (252.0 mg, 3.96 mmoles) was refluxed for 3 days. After cooling to room temperature, the mixture was poured into ice water (150 ml) with stirring. The product was extracted with chloroform (30 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solvent was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing the product were combined and then evaporated under reduced pressure. Cold water (100 ml) was added to the residue with stirring. The resulting crystals were filtered and dried in air to give compound **11a** (106.8 mg, 22%) as colorless needles.

3-(Dimethylamino)-6-(4-cyanophenoxy)pyridazine (**11b**).

A mixture of **10b** (500 mg, 2.16 mmoles) [10] in *N,N*-dimethylformamide (10 ml) in the presence of Cu powder (300.0 mg, 4.72 mmoles) was refluxed for 3 days. After cooling to room temperature, the mixture was poured into ice water (200 ml) with stirring. The product was extracted with chloroform (30 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solution was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing the product were combined and then evaporated under reduced pressure. Cold water (100 ml) was added to the residue with stirring. The resulting crystals were filtered and dried in air to give compound **11b** (77 mg, 14%) as colorless needles.

3-(Dimethylamino)-6-(4-nitrophenoxy)pyridazine (**11c**).

A mixture of **10c** (500 mg, 2.47 mmoles) [10] in *N,N*-dimethylformamide (10 ml) in the presence of Cu powder (313.7 mg, 4.94 mmoles) was refluxed for 37 hours. After cooling to room temperature, the mixture was poured into ice water (150 ml) with stirring. The product was extracted with chloroform (30 ml x 2), and the solution was then dried over anhydrous magnesium sulfate. The organic solution was evaporated under reduced pressure. The resulting residue was applied to the top of an open-bed silica gel column (2.5 x 5 cm). The column was eluted with chloroform. Fractions containing the product were combined and then evaporated under reduced pressure. Cold water (100 ml) was added to the residue with stirring. The resulting crystals were filtered and dried in air to give compound **11c** (231 mg, 36%) as yellow needles.

4-Chloro-5-(dimethylamino)-2-methyl-2*H*-pyridazin-3-one (**12**).

A mixture of **2** (110 mg, 0.63 mmole), methyl iodide (117.7 μl, 1.89 mmoles), potassium carbonate (174.1 mg, 1.26 mmoles), and acetone (5 ml) was refluxed for 10 hours. The mixture was filtered using a glass filter and washed with ethyl acetate. The combined organic solvent was evaporated under reduced pressure to give the residue, which was purified by column chromatography on silica gel (elution solvent: ethyl acetate) to afford **12** (79 mg, 67%) as colorless prisms.

5-(Dimethylamino)-2-methyl-2*H*-pyridazin-3-one (**13**).

A mixture of **12** (90 mg, 0.48 mmole), Pd/C (50 mg), aqueous sodium hydroxide (10%, 1 ml) and ethanol (5 ml) was stirred for 30 minutes under a hydrogen atmosphere (using a toy balloon) at room temperature. After removal of the catalyst by filtration, the residue was washed with ethanol. The resulting solution was evaporated under reduced pressure. The residue was dissolved in ethyl acetate and the solution was dried over anhydrous magnesium sulfate. The combined organic solvent was evaporated under reduced pressure to give **13** (27 mg, 37%) as colorless prisms.

3,5-Di-(dimethylamino)pyridazine (**14**).

A mixture of Pd/C (20 mg), **9** (93 mg, 0.40 mmole), and methanol (5 ml) was stirred for 24 hours under a hydrogen atmosphere (using a toy balloon) at room temperature. After removal of the catalyst by filtration using Celite 545, the residue was washed with methanol (5 ml x 2). The solution was evaporated under reduced pressure to give **14** (58 mg, 87%) as colorless prisms.

Acknowledgment.

This work was supported by the Brain Korea 21 Project.

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