Halogenation and Nitration of Naphtho[1,2-*c*][1,2,5]thiadiazole

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Bromination of naphtho[1,2-*c*][1,2,5]thiadiazole (**I**) gives either an addition product, 4,5-dibromo-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**II**), or a substitution product, 5,6-dibromonaphtho[1,2-*c*][1,2,5]thiadiazole (**III**), depending on the reaction conditions. Dehydrobromination of **II** gives 5-bromonaphtho[1,2-*c*][1,2,5]thiadiazole (**IV**). Chlorination of **I** gives the corresponding addition product **V** or 5-chloronaphtho[1,2-*c*][1,2,5]thiadiazole (**VI**). Compound **VI** can also be obtained by dehydrochlorination of **V**. The nitration of **I** produces a mixture of isomeric substitution products, 9-nitro **VIII** and 6-nitronaphtho[1,2-*c*][1,2,5]thiadiazoles (**VII**).

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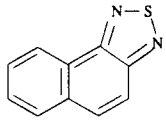
Naphtho[1,2-*c*][1,2,5]thiadiazole (**I**) may be considered to be a heterocyclic analog of phenanthrene and it is therefore of interest to compare the bromination, chlorination and nitration of **I** with phenanthrene. Phenanthrene is reported to react with chlorine in acetic acid to give a mixture of *cis*- and *trans*-9,10-dichloro-9,10-dihydrophenanthrene (42%) with the *cis* isomer predominating, 9-acetoxy-10-chloro-9,10-dihydrophenanthrene (14%) with the *trans* isomer predominating and 9-chlorophenanthrene (34%). While neither the *cis* or *trans*-9,10-dichlorophenanthrenes were converted in significant amounts to the 9-chlorophenanthrene under the experimental conditions, both could be converted on treatment with alcoholic base [1]. Bromination of phenanthrene gives either the 9,10-addition product [2,3] or substitution at the 9-position [4]. The addition of bromine in acetic acid at 25° is reported to be nearly second order due to the predominance of addition over substitution [2]. A major product from addition of bromine to phenanthrene in methanol has been assigned the *trans* structure on the basis of dipole moment and nmr spectrum [5].

Phenanthrene, present in large excess, has been reported to react with nitric acid-acetic anhydride to give mixtures of 1-nitro (26%), 3-nitro (22%) and 9-nitrophenanthrene (35%) [6]. The nitration of a compound closely related to **I**, naphtho[1,2-*c*][1,2,5]oxadiazole, has been previously reported [7,8,9]. Degradative evidence obtained by Green and Rowe [7] indicate that the oxadiazole is nitrated in the 6 or 9 position under conditions favoring mononitration. Bogdanov and Petrov reported the formation of the 8 and 6 isomers in a 3:2 ratio [8] and that nitration of the naphtho[1,2-*c*][1,2,5]oxadiazole bisulfite compound gave the 6-isomer [9].

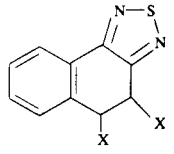
An earlier study [10] of the bromination and chlorination of **I** reported that when molten **I** was treated with either bromine or chlorine, addition of two halogens took place to give respectively **II** or **V**. We have found that bromination of molten **I** gives a disubstitution product rather than an addition product and have assigned this substitution product the structure 5,6-dibromonaphtho[1,2-*c*]-[1,2,5]thiadiazole (**III**) on the basis of its nmr spectrum and elemental analysis. The nmr spectrum showed a total of four protons located in the aromatic region of the spectrum. Other conditions used by us which lead to the formation of **III** include treatment with bromine vapor according to the method of Buckles and Wheeler for the bromination of phenanthrene [4], and treatment with a four-fold excess of bromine at room temperature in the presence of zinc chloride.

When the bromination of **I** is carried out in glacial acetic acid at 49°, the addition product 4,5-dibromo-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**II**), mp 135-137° is formed. The structure of **II** was confirmed by its nmr spectrum (4,5-protons) and by its conversion *via* dehydrohalogenation to 5-bromonaphtho[1,2-*c*][1,2,5]thiadiazole (**IV**). Compound **II** (mp 135-137°) is probably the same as that obtained by Pesin *et al.* (mp 132-132.5°), X'_1,X'_2-dibromonaphtho[1,2-*c*][1,2,5]thiadiazole. Addition of bromine rather than substitution to produce **II** also occurs under the following conditions: in acetic acid at room temperature, in refluxing carbon tetrachloride, and with a slight excess of bromine in no solvent at room temperature in the presence of either zinc chloride or aluminum chloride. In a repetition of this last experiment, but without the zinc chloride, the same product is obtained but in lower yield.

We found that chlorination of molten **I** gave 4,5-dichloro-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**V**) in agreement with results obtained by Pesin *et al.* These earlier workers also reported that chlorination in acetic acid gave an X'_2,X'_2-dichloronaphtho[1,2-*c*][1,2,5]thiadiazole. When we used these same conditions we obtained not a substitu-



I



II, X = Br

V, X = Cl

tion product but the same addition product **V** that we obtained by chlorination of molten **I**. We found that **V** could also be obtained by chlorination of molten **I** in the presence of iron powder, and reaction of **I** with liquid chlorine or sulfuryl chloride. The nmr spectrum of **V** is consistent with the structure assigned.

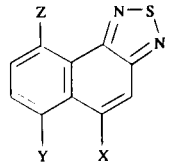
From the chlorination of molten **I** in the presence of zinc chloride we obtained 5-chloronaphtho[1,2-*c*][1,2,5]-thiadiazole (**VI**) reported by Pesin *et al.* for the compound they made from 4-chloro-1,2-diaminonaphthalene. Chlorination of **I** with *N*-chlorosuccinimide in sulfuric acid, a procedure known to give aromatic chlorination [11], also gave **VI**.

Under several different nitration reaction conditions including various concentrations of nitric acid, we find that **I** gives a mixture of the 6-isomer **VII** and the 9-isomer **VIII**. Structural assignments were made on the basis of the nmr spectra. The ratio of the 6 to 9-isomer as determined by gas chromatography was essentially 3:2 regardless of the nitrating conditions we tried. Separation of the two isomers by recrystallization was quite difficult. Recrystallization from acetone was preferable to recrystallization from methanol for purification of the 6-isomer, but it was necessary to use gas chromatography to obtain an analytical sample. To obtain pure 9-isomer, advantage was taken of the preferential reduction of the 6-isomer by ammonium sulfide. This resulted in a mixture of the 6-amino compound and the 9-nitro compound. The latter could then be recrystallized from methanol to remove the more soluble amino compound. Preparation of the 6-amino compound could also be accomplished by catalytic reduction of the 6-nitro isomer.

The preparation of some acylated 6-aminonaphtho-[1,2-*c*][1,2,5]thiadiazoles, **X-XI**, as well as of 7-nitro-6-acetamidonaphtho[1,2-*c*][1,2,5]thiadiazole is described in the Experimental.

In summary, it appears that **I** on halogenation behaves much like phenanthrene in that it can undergo either addition or substitution at the middle ring. Phenanthrene in acetic acid gives addition along with some substitution. Similarly, **I** in acetic acid also gives addition products in both chlorination and bromination reactions. Chlorination of molten **I** also results in addition; if zinc chloride is present under these conditions monosubstitution to produce **VI** occurs. Reaction of **I** with *N*-chlorosuccinimide in 50% sulfuric acid produces monosubstitution as expected. The bromination reaction products formed in the presence of zinc chloride depend on the concentration of bromine. With *ca.* equimolar amounts of bromine and **I**, addition to give **II** is observed while at higher concentrations disubstitution to give **III** occurs. Bromination under the conditions of Buckles and Wheeler also resulted in disubstitution of **I**.

In contrast to the halogenation of **I**, the nitration results show that **I** behaves more like 1,2-dinitronaphthalene than like phenanthrene. It has been reported that nitration of 1,2-dinitronaphthalene gives 1,2,5-trinitro and 1,2,8-trinitronaphthalenes in the ratio of 2:2.6 [12]. In our work, the nitration of **I** produces a mixture of **VII** and an unidentified compound which is probably **VIII**. These results would be expected since the protonated heterocyclic ring behaves as a *meta* directing or deactivating group and also appear to be in agreement with the results of Pesin and Kaukhova [13].



III, Z = H, Y = Br, X = Br

IV, Z = H, Y = H, X = Br

VI, Z = H, Y = H, X = Cl

VII, Z = H, Y = NO_2, X = H

VIII, Z = NO_2, Y = H, X = H

IX, Z = H, Y = NH_2, X = H

X, Z = H, Y = CH_3-C(=O)-NH, X = H

XI, Z = H, Y = CH_3-CH_2-C(=O)-NH-, X = H

EXPERIMENTAL

All melting points were determined on a Fisher-Johns apparatus and are uncorrected. Infrared spectra were obtained on a Beckman Model IR-8 spectrophotometer and ultraviolet spectra on a Perkin-Elmer Model 202 spectrophotometer. Nuclear magnetic resonance spectra were determined on a Varian HA-60-IL spectrometer with a C-1024 time averaging computer using tetramethylsilane as an internal standard. Gas chromatographic analyses were determined using a 20% SE-30-Chromosorb P column. Elemental analyses were determined by Atlantic Microlab, Inc. or on an F and M Model 185 C,H,N-Analyzer by Mr. Daryl Sharp.

4,5-Dibromo-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**II**).

To a stirred solution of naphtho[1,2-*c*][1,2,5]thiadiazole (**I**) (0.25 g, 1.4 mmoles) in 10 ml of acetic acid was added a solution of bromine (0.1 ml, 1.8 mmoles) in 2 ml of acetic acid. The mixture was stirred at room temperature for 2 hours and then evaporated to give 0.37 g, mp 65-75°. The dibromodihydro compound was separated from the starting material on the basis of its lesser solubility in methanol. The solid was taken up in methanol, heated slightly on a steam bath and filtered. The filtrate contained 0.20 g (80% recovery) of starting material, mp 72-74°, mmp 72-74°. The insoluble material was recrystallized 6 times from methanol to give 0.06 g (13%) of 4,5-dibromo-4,5-dihydronaphtho[1,2-*c*]-[1,2,5]thiadiazole (**II**), mp 135-137°; ^{1}H nmr (deuteriochloroform): δ 8.20 and 7.54 (m and d, 4 H of aromatic ring), 5.97 and 5.72 (doublets, 2 H at C-4 and C-5, J = 2.5); uv (95% ethanol): λ max, nm (ϵ), 210 (17,150), 226 (18,850), 300

(14,750).

Anal. Calcd. for $C_{10}H_6N_2SBr_2$: C, 34.70; H, 1.73; N, 8.09. Found: C, 34.68; H, 1.72; N, 7.99.

When the above reaction was carried out using 3 times as much bromine at 49° for 4 hours, 60% of starting material was recovered and a 15% yield of **II** was obtained.

Bromination in refluxing carbon tetrachloride for 1 hour, with 650% molar excess bromine gave 26% of **II** and 64% recovery of starting material.

Bromination of **I** (1.4 mmoles) in 1.8 mmoles bromine containing 0.1 g of zinc chloride for 1 hour at room temperature gave a 36% yield of **II**.

5,6-Dibromonaphtho[1,2-*c*][1,2,5]thiadiazole (**III**).

To molten **I** (0.25 g, 1.4 mmoles) heated on a steam bath was added bromine (0.2 ml, 3.7 mmoles). After 1 hour, the bromine was evaporated and the residue was taken up in acetone, treated with Norit and cooled to give 0.2 g (43%) which melted at 212-215° after 6 recrystallizations (acetone); ^{1}H nmr (deuteriochloroform): δ 8.68 (d, 1 H at C-9), 8.63 (s, 1 H at C-4), 7.97 (2d, 1 H at C-7), 7.56 (t, 1 H at C-8); uv (chloroform): λ max, nm (ε), 272 (31,100), 277 (38,600), 331 (5,500), 356 (8,310), 373 (7,900).

Anal. Calcd. for $C_{10}H_4N_2SBr_2$: C, 34.90; H, 1.16, N, 8.14. Found: C, 34.52; H, 1.01; N, 7.98.

Bromination of **I** (1.4 mmoles) in 0.3 ml of bromine (5.5 mmoles) containing 0.10 g of zinc chloride at room temperature for 1.6 hours gave 0.23 g (49%) of **III** which melted at 212-215° after 5 recrystallizations from acetone. Repetition of this experiment without the addition of zinc chloride gave a 30% yield of crude **III**.

Bromination of **I** by the method of Buckles and Wheeler [4] using 1.4 mmoles of **I** and 28 mmoles of bromine gave **III** (71%) mp 160-195°, mp 212-214° after five recrystallizations from acetone.

5-Bromonaphtho[1,2-*c*][1,2,5]thiadiazole (**IV**).

A solution of 4,5-dibromo-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**II**) and sodium hydroxide (0.20 g, 0.6 mmole) in 0.4 ml of water and 10 ml of 95% ethanol was refluxed for 1 hour, diluted with 15 ml of water, cooled and filtered to give 0.14 g (93%) of 5-bromonaphtho[1,2-*c*][1,2,5]thiadiazole (**IV**), mp 120-126°, mp after 2 recrystallizations from 50% acetic acid, 128-129°; lit mp 124-125° [6]; mmp with starting material 107-115°; ^{1}H nmr (deuteriochloroform): δ 8.68 (m, 1 H at C-9), 8.10 (s, 1 H at C-4), 7.71 (m, 3 H at C-6, C-7, and C-8); uv (1,4-dioxane): λ max, nm (ε), 271 (26,320), 278 (39,740), 320 sh (5,510), 331 (6,240), 349 (7,220), 365 (6,770).

Anal. Calcd. for $C_{10}H_5N_2SBr$: C, 45.25; H, 1.89; N, 10.57. Found: C, 45.26; H, 1.96; N, 10.59.

4,5-Dichloro-4,5-dihydronaphtho[1,2-*c*][1,2,5]thiadiazole (**V**).

Molten **I** (0.25 g, 1.4 mmoles) was saturated with chlorine for 5 minutes while heated on the steam bath. The liquid formed a solid product almost immediately. The residue was taken up in methanol, treated with Norit and cooled to give 0.32 g (93%) of **V** which melted at 153-154° after 5 recrystallizations from methanol, lit mp 153-153.5° [6]; ^{1}H nmr (deuteriochloroform): δ 8.04 and 7.38 (2m, 4 H of aromatic ring), 5.53 and 5.31 (2d, J = 2.5, 2 H at C-4 and C-5); uv (95% ethanol): λ max, nm (ε), 218 (19,800), 257 (4,610), 297 (15,200), 308 sh (12,080).

Anal. Calcd. for $C_{10}H_6N_2SCl_2$: C, 46.65; H, 2.34; N, 10.90. Found: C, 46.54; H, 2.35; N, 10.79.

Chlorination of molten **I** (0.25 g, 1.4 mmoles) (steam bath) with 10 ml of sulfuryl chloride, 1 hour gave 0.23 g **V** (68%), mp 153-154° after 5 recrystallizations from methanol.

Chlorination of **I** (0.50 g, 2.7 mmoles) in 25 ml of acetic acid saturated with chlorine gas for 15 minutes at 48-50° gave 0.48 g (70%) of **V**, mp 149-150° after 5 recrystallizations from methanol. (Pesin *et al.* [6] reported that under these reaction conditions a compound mp 138-140° was obtained and to which they assigned the structure X_1',X_2'-dichloronaphtho[1,2-*c*][1,2,5]thiadiazole. Their structure is inconsistent with the nmr spectrum and dehydrohalogenation behavior).

Chlorination of **I** (0.25 g, 1.4 mmoles) in the presence of 0.10 g of iron filings (20 mesh) by adding chlorine gas for 1 hour at room temperature gave 0.26 g of **V**, after 5 recrystallizations the mp was 151-152°, mmp 151-153° with an authentic sample.

Chlorination of **I** (0.50 g, 2.8 mmoles) in an excess of liquid chlorine, followed by gradual warming (24 hours) to room temperature gave 0.53 g of **V**, mp 152-153° after 5 recrystallizations.

5-Chloronaphtho[1,2-*c*][1,2,5]thiadiazole (**VI**).

Compound **I** (0.25 g, 1.4 mmoles) and 0.1 g of zinc chloride was heated on the steam bath while chlorine gas was passed in for 15 minutes. The cooled mixture was taken up in acetone and chromatographed on alumina using 200 ml of hexane as the eluting solvent. Recrystallization from ethanol-water gave 0.18 g, mp 81-90°. Five additional recrystallizations gave 0.15 g, (50%) of **VI**, mp 117-119°. A sample collected by gas chromatography melted at 121-123°, lit mp 124° [6].

Chlorination of **I** (0.25 g, 1.4 mmoles) in 10 ml of concentrated sulfuric acid and 10 ml of water by the addition of *N*-chlorosuccinimide (0.19 g, 1.4 mmoles) and heating at 65-70° for 2 hours gave 0.17 g (57%) of **VI**, mp 100-112°. Gas chromatography gave mp 120-122°, mmp 120-122° with sample prepared above by chlorination in the presence of zinc chloride.

Preparation of **VI** by Dehydrochlorination of **V**.

A mixture of **V** (0.20 g, 0.78 mmole), 10 ml of 95% ethanol, and 0.20 g (5 mmoles) of sodium hydroxide in 0.4 ml of water was refluxed 1 hour, cooled, diluted with 10 ml of water, heated 15 minutes, and cooled to give 0.14 g (82%) of **VI**, mp 118-123°. After two recrystallizations from dilute alcohol, mp 121-123°, mixture mp with sample of **VI** prepared by direct chlorination of **I** in presence of zinc chloride, 120-123°; ^{1}H nmr (deuteriochloroform): δ 8.37 (m, 1 H at C-9), 7.75 (s, 1 H at C-4), 7.62 (m, 3 H at C-6, 7, 8); uv (1,4-dioxane): λ max, nm (ε), 271 (25,950), 277 (40,810), 319 sh (4,830), 331 (5,890), 349 (7,070), 365 (6,590).

6-Nitronaphtho[1,2-*c*][1,2,5]thiadiazole (**VII**).

To naphtho[1,2-*c*][1,2,5]thiadiazole (0.25 g, 1.4 mmoles) and 5 ml of concentrated sulfuric acid cooled in an ice bath was added a solution of 0.4 ml of nitric acid (d = 1.5) in 3 ml of concentrated sulfuric acid. The mixture was stirred for 0.5 hour and then poured over 100 g of ice-water. The solid was removed by filtration, dried, taken up in benzene and analyzed by gas chromatography at 215°. The chromatography showed two peaks, retention times 12 and 17 minutes, relative areas 62:38. In several other experiments using either 0.4 or 0.1 ml of nitric acid of either 1.5, 1.42 or 1.1 density gave essentially the same ratio of products. Component 1, purified by gas chromatography, mp

184-186° was identified as 6-nitronaphtho[1,2-*c*][1,2,5]thiadiazole on the basis of its ^{1}H nmr (in a mixture of carbon tetrachloride-carbon disulfide at 55°): δ 8.87 (d, 1 H at C-7), 8.33 and 7.83 (2 d's, 2 H at C-4,5), 8.08 (q, 1 H at C-9), 7.60 (t, 1 H at C-8); uv (95% ethanol): λ max, nm (ε), 220 (30,200), 236 (13,700), 293 (18,000), 323 (8,150), 339 (8,850), 355 (7,490). Component 2 could not be completely purified, but is presumably the 9-isomer.

Anal. Calcd. for $C_{10}H_5N_3O_2S$: C, 51.94; H, 2.18; N, 18.18. Found: C, 51.67; H, 2.12; N, 17.98.

6-Aminonaphtho[1,2-*c*][1,2,5]thiadiazole (**IX**).

Hydrogenation of 6-nitronaphtho[1,2-*c*][1,2,5]thiadiazole (**VII**) (0.28 g, 1.2 mmoles) in 200 ml of absolute ethanol, 3 ml of acetic acid with 0.3 g of 5% Pd/C for 8 hours gave **IV** mp 150-152° (methanol); ^{1}H nmr (deuteriochloroform): δ 8.18 (d, 1 H at C-7), 7.85 and 7.71 (2 d's, 2 H at C-4,5), 7.47 (t, 1 H at C-8), 6.97 (d, 1 H at C-9), 3.50 (broad, 2 H of NH_2); uv (95% ethanol) λ max, nm (ε), 208 (19,500), 238 (32,600), 278 (17,450), 409 (5,550).

Anal. Calcd. for $C_{10}H_7N_3S$: C, 59.68; H, 3.51; N, 20.88. Found: C, 59.64; H, 3.40; N, 20.91.

6-Acetamidonaphtho[1,2-*c*][1,2,5]thiadiazole (**X**).

Acetylation of **IX** with acetic anhydride gave **V**, mp 236-238° (methanol); ^{1}H nmr (deuteriochloroform-deuterioacetone at 60°): δ 8.85 (d, 1 H at C-7), 8.00 and 7.53 (2 d's, 2 H at C-4,5), 7.86 (d, 1 H at C-9), 7.58 (t, 1 H at C-8), 2.22 (s, 3 H of CH_3); uv (95% ethanol): λ max, nm (ε), 210 (22,300), 226 (31,650), 271 sh (27,600), 276 (32,000), 345 (5,700), 361 (6,900).

Anal. Calcd. for $C_{12}H_9N_3OS$: C, 59.24; H, 3.73; N, 17.28. Found: C, 59.42; H, 3.73; N, 17.72.

6-Propionamidonaphtho[1,2-*c*][1,2,5]thiadiazole (**XI**).

Acylation of **IX** with propionic anhydride gave **XI**, mp 203-204° (methanol); ^{1}H nmr (deuteriochloroform): δ 8.45 (d, 1 H at C-7), 7.58 (m, 4 H at C-4,5,8,9), 2.50 (q, 2 H of CH_2), 1.24 (t, 3 H of CH_3); uv (95% ethanol): λ max, nm (ε), 212 (22,680), 229 (32,680), 272 sh (27,990), 278 (33,270), 349 sh (6,220), 364 (7,240).

Anal. Calcd. for $C_{13}H_{11}N_3OS$: C, 60.68; H, 4.31; N, 16.33. Found: C, 60.75; H, 4.22; N, 16.34.

7-Nitro-6-acetamidonaphtho[1,2-*c*][1,2,5]thiadiazole.

Nitration of **X** with nitric-sulfuric acids at 0° gave 7-nitro-6-acetamidonaphtho[1,2-*c*][1,2,5]thiadiazole, mp 275-278° (methanol); the ^{1}H nmr showed a 2:2:1:3 ratio of peaks. The aromatic portion appears to be two AB patterns with the B portions overlapping. There were peaks at 7.75 (NH) and 2.82 (CH_3); uv (95% ethanol): λ max, nm (ε), 209 (25,790), 228 (28,410), 278 (29,780), 348 sh (5,610), 361 (6,110).

Anal. Calcd. for $C_{12}H_8N_4O_3S$: C, 49.99; H, 2.80; N, 19.44. Found: C, 50.05; H, 2.68; N, 19.33.

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