of 1,3-dichloro-2-propanone, and 0.3 mL of triethylamine in 10 mL of acetone. After the reaction mixture had been evaporated, the remaining residue was mixed with water. The insoluble oily material was separated from water, and the oil was solidified with acetone, giving crude **3g**: 0.58 g (74%); mp 196–197 °C. Recrystallization from DMF–methanol provided 0.42 g (55%) of **3g** as a yellow solid: mp 196–197 °C; IR (KBr) 3400, 3300 (NH) and 1620 cm^{-1} (C=O); NMR (Me_2SO-d_6) δ 1.40 (t, 6 H, J = 7 Hz, CH_3), 4.40 (q, 4 H, J = 7 Hz, CH_2), 7.67 (s, 4 H, NH_2); MS, m/e 314 (M^+).

Registry No. 1, 29422-34-2; **2a**, 86690-06-4; **3a**, 86690-07-5; **3b**, 86690-08-6; **3c**, 86690-09-7; **3d**, 86690-10-0; **3e**, 86690-11-1; **3f**, 86690-12-2; **3g**, 86695-78-5; $PhCOCH_2Br$, 70-11-1; 4-$ClC_6H_4COCH_2Br$, 536-38-9; 4-$BrC_6H_4COCH_2Br$, 99-73-0; 4-$MeOC_6H_4COCH_2Br$, 2632-13-5; 4-$PhC_6H_4COCH_2Br$, 135-73-9; $MeCOCH_2Br$, 598-31-2; 1,3-dichloro-2-propane, 534-07-6.

Synthesis and Rearrangements of Dihydro-1,4-oxazepine and Dihydro-1,4-thiazepine Derivatives

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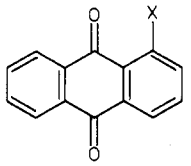
Received February 2, 1983

In a drug development program dealing with a study of the structure–activity relationship of substituted anthraquinones (anthracene-9,10-diones) as antineoplastic agents, we wished to prepare 1-(2-aminoethoxy)anthraquinone (**1a**) and the corresponding sulfur analogue **1b** for evaluation of their antitumor activity.[1] Our efforts in this research have uncovered some interesting heterocylic chemistry which we report.

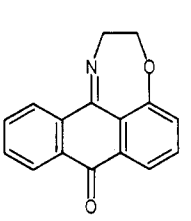


1a, X = $OCH_2CH_2NH_2$
b, X = $SCH_2CH_2NH_2$
c, X = Cl
d, X = $OCH_2CH_2N(CH_3)_2$
e, X = $NHCH_2CH_2OH$
f, X = F
g, X = $OCH_2CH_2N{=}CHC_6H_5$
h, X = $OCH_2CH_2NH_2 \cdot HCl$

It had previously been reported by Cheng and coworkers[1] that treatment of 1-chloroanthraquinone (**1c**) with the potassium salt of 2-(dimethylamino)ethanol yielded **1d** (60%, isolated as the hydrochloride salt). Our initial attempt to prepare **1a** was to react **1c** with 1 equiv of the sodium salt of 2-aminoethanol in Me_2SO as solvent (65 °C, 15 h). However, this reaction led to **1e** (8%) along with starting anthraquinone **1c** (56%). Compound **1e** could also be prepared in a 73% yield by treatment of **1c** with 5 equiv of 2-aminoethanol in Me_2SO at 70 °C for 20 h. Since displacements by amines are known to proceed more readily with 1-fluoroanthraquinone (**1f**)[2] than **1c** the preparation of **1e** could most readily be accomplished by treatment of **1f** with excess 2-aminoethanol at 25 °C for 16 h (68% yield).

To preclude reaction at the nitrogen atom, we converted 2-aminoethanol to the corresponding imine[3] by treatment with benzaldehyde in a benzene solution with azeotropic removal of water. Addition of **1f** to the sodium salt of this imine (treatment with NaH in THF) followed by stirring at room temperature for 1.5 h leads to **1g** (90%). The imine **1g** could be readily converted into the hydrochloride salt **1h** (96%) by hydrolysis in dilute HCl. The free amine could be obtained by treatment of **1h** with aqueous sodium bicarbonate.

Some interesting chemistry was uncovered during the purification of the imine **1g**. Column chromatography of the crude reaction mixture on silica gel led to a new product which was identified as the dihydro-1,4-oxazepine derivative **2** (68%) by ^1H and ^{13}C NMR analysis. The conversion of pure **1g** to the dihydro-1,4-oxazepine **2** (93%) could be most readily accomplished by merely passing a solution of the imine in CH_2Cl_2 through a column of silica gel. The dihydro-1,4-oxazepine derivative **2** arises from initial hydrolysis of the imine linkage to the amine **1a** and subsequent intramolecular condensation of **1a** on the silica gel.



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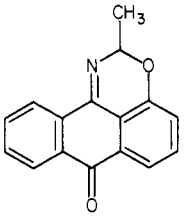


3

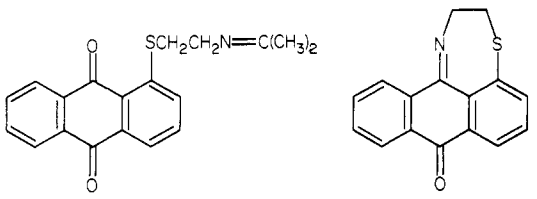
An unexpected rearrangement was discovered when **1a** was refluxed in toluene. After 20 h of refluxing, in addition to the expected dihydro-1,4-oxazepine derivative **2** (56%), a significant amount of the rearranged 1,3-oxazine derivative **3** (36%) was formed. It was then found that merely refluxing the amine **1a** or the imine **1g** in glacial acetic acid for 1 h also led to **3** (68% and 67% yields, respectively). In addition to the 1,3-oxazine **3**, both reactions produce 1-hydroxyanthraquinone in approximately 20% yields. This latter product arises from hydrolysis of the oxazine **3** since on heating **3** for 24–48 h in glacial acetic acid a nearly quantitative yield of 1-hydroxyanthraquinone was obtained (TLC assay).

The sulfur analogue **1b** was prepared by treatment of 2-aminoethanethiol hydrochloride in a basic medium of dioxane–water–ethanol with **1f**. The thio amine **1b** was isolated and characterized as its Schiff base **4** which formed upon crystallization of **1b** from acetone. When **1b** was refluxed in toluene (20 h) followed by silica gel chromatography, the thiazepine derivative **5** was isolated in a 56% yield. The crude base **1b** on being refluxed in glacial acetic acid for 1 h led to 7*H*-dibenzo[*de*,*h*]quinolin-7-one (**6**, 66%), the product of a formal loss of H_2S. This result contrasts markedly with the transformation undergone by the oxazepine derivative **2**.

It has previously been reported that treatment of **1c** with *o*-aminothiophenol followed by treatment of the product with refluxing acetic acid leads to the thiazepine derivative **7**. However, this thiazepine **7** on being refluxed in diethyl phthalate leads to **8**, the product of a formal extrusion of sulfur.[4]

(1) Zee Cheng, R. K.-Y.; Podrebarac, E. G.; Menon, C. S.; Cheng, C. C. *J. Med. Chem.* **1979**, *22*, 501.
(2) (a) Solodar, W. E.; Simon, M. S. *J. Org. Chem.* **1962**, *27*, 689. (b) Shein, S. M.; Shternshis, M. V. *J. Org. Chem. USSR (Engl. Transl.)* **1971**, *7*, 1278 and references cited therein.
(3) Bergman, E. D.; Zimkin, E.; Pinchas, S. *Recl. Trav. Chim. Pays-Bas* **1952**, *71*, 168.

0022-3263/83/1948-3341$01.50/0



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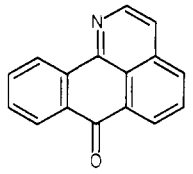
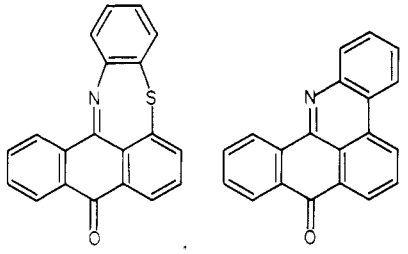
5



6



7 8

While mechanistic rationalizations of the rearrangements reported above are tempting, perhaps speculation should await the results of further studies.

Experimental Section

Melting points were determined on a Thomas-Hoover melting point apparatus and are uncorrected. ^{1}H NMR and ^{13}C NMR were run on a Bruker WM-250 pulsed Fourier transform NMR spectrometer. TLC precoated silica gel plates (Eastman chromagram sheets with fluorescent indicator) were used to monitor reactions. For column chromatography Baker analyzed 80–200-mesh silica gel was utilized. Microanalyses were performed by Robertson Laboratories, Florham Park, NJ. Mass spectra were run on a Finnigan MAT 4610 mass spectrometer.

1-[(2-Hydroxyethyl)amino]anthracene-9,10-dione (1e). (A) To a solution of 2-aminoethanol (1.5 g, 0.0245 mol) in Me_2SO (25 mL) was added 1-chloroanthraquinone (1.22 g, 0.0050 mol). The solution was heated at 70 °C with stirring for 20 h under a nitrogen atmosphere. The reaction mixture was cooled and poured into ice–water (150 mL), and the precipitate was collected by filtration, washed with water, and dried in vacuo to yield a red solid (1.37 g). Recrystallization from a CH_2Cl_2–pentane mixture afforded dark red needles of **1e** (0.98 g, 73%) in three crops: mp 170–172 °C (lit.[5] mp 170–171 °C); ^{1}H NMR (Me_2SO-d_6) δ 10.8 (br t, 1 H), 7.3–8.3 (m, 7 H), 5.0 (t, 1 H), 3.7 (m, 2 H), 3.4 (m, 2 H).

(B) Excess 2-aminoethanol (8.0 mL, 0.133 mol) and 1-fluoroanthraquinone[6] (0.30 g, 0.001 33 mol) were stirred at room temperature for 16 h under a nitrogen atmosphere. The reaction mixture was poured into ice–water (50 mL), and the precipitate was filtered, washed with water, and dried in vacuo to yield a red solid (0.295 g). Recrystallization from methanol afforded dark red needles of **1e** (0.24 g, 68%) in three crops.

1-[2-(Benzylideneamino)ethoxy]anthracene-9,10-dione (1g). 2-(Benzylideneamino)ethanol[3] (2.5 g, 0.0168 mol) was added to a suspension of NaH (60% oil dispersion, 0.49 g, 0.0123 mol) in THF (15 mL) under a nitrogen atmosphere. After the mixture was stirred for 0.5 h, 1-fluoroanthraquinone (2.5 g, 0.0111 mol) was added, and the flask was washed down with THF (10 mL). The reaction mixture was followed to stir for 1.5 h at room temperature. The mixture was poured into ice–water (75 mL), and the resulting precipitate was filtered, washed with water, and dried in vacuo to give a yellow solid (3.53 g, 90%). Recrystallization of 1.0 g from ethanol gave yellow needles: 0.86 g; mp 156–158 °C; ^{1}H NMR ($CDCl_3$) δ 8.6 (s, 1 H), 7.3–8.3 (m, 12 H), 4.5 (t, 2 H), 4.2 (t, 2 H); ^{13}C NMR ($CDCl_3$) δ 183.49, 181.99, 164.86, 159.85, 159.82, 136.28, 135.75, 135.20, 134.78, 134.12, 133.08, 132.59, 130.75, 128.56, 128.33, 127.11, 126.58, 120.07, 119.96, 69.18, 59.99; MS, *m/e* (relative intensity) 355 (29.6, M^+), 259 (28.3), 248 (27.4), 236 (42.7), 224 (100), 223 (31.7), 149 (37.7), 69 (42.4), 55 (46.8). Anal. Calcd for $C_{23}H_{17}NO_3$: C, 77.73; H, 4.82; N, 3.94. Found: C, 77.53; H, 4.76; N, 4.04.

1-(2-Aminoethoxy)anthracene-9,10-dione Hydrochloride (1h). Imine **1g** (1.0 g, 0.002 82 mol) was stirred in 10% HCl (20 mL) at 75 °C for 1 h. The cooled reaction mixture was filtered and the collected solid dried in vacuo to give a bright yellow solid (0.82 g, 96%). Crystallization from an ethanol–methanol mixture yielded yellow needles: mp 213–216 °C dec; ^{1}H NMR (Me_2SO-d_6) δ 8.4 (br s, 3 H), 8.1–8.2 (m, 2 H), 7.8–8.0 (m, 4 H), 7.6–7.75 (m, 1 H), 4.44 (t, 2 H), 3.2–3.4 (m, 2 H). Anal. Calcd for $C_{16}H_{14}NO_3Cl$: C, 63.27; H, 4.65. Found: C, 63.15; H, 4.89.

2,3-Dihydro-8*H*-anthra[9,1-*ef*][1,4]oxazepin-8-one (2). The Schiff base **1g** (1.0 g, 0.002 82 mol) was hydrolyzed by chromatography on silica gel. The first fraction was eluted with CH_2Cl_2 followed by elution of the major fraction with methanol. After evaporation of the methanol, a yellow-orange solid (0.653 g, 93%) remained. Crystallization from ethanol afforded **2** as orange needles: mp 136–137 °C; ^{1}H NMR ($CDCl_3$) δ 7.3–8.7 (m, 7 H), 4.55 (m, 2 H), 4.4 (m, 2 H); ^{13}CNMR δ 183.59, 157.91, 157.60, 139.06, 134.62, 133.51, 131.91, 131.28, 130.31, 126.45, 126.37, 126.30, 121.87, 119.73, 73.28, 55.30; MS, *m/e* (relative intensity) 249 (60.7, M^+), 248 (100.0), 220 (40.7), 164 (65.1), 163 (73.5). Anal. Calcd for $C_{16}H_{11}NO_2$: C, 77.10; H, 4.45; N, 5.62. Found: C, 77.10; H, 4.45; N, 5.47.

2-Methylanthra[9,1-*de*][1,3]oxazin-7(2*H*)-one (3). (A) Preparation from 1a. The hydrochloride salt **1h** was added to an aqueous sodium bicarbonate solution, and the mixture was extracted with CH_2Cl_2. The solvent was removed under reduced pressure to yield the free amine **1a** as a yellow-orange solid. A solution of **1a** (0.080 g, 0.000 30 mol) in glacial acetic acid (5.0 mL) was refluxed with stirring for 1 h. The cooled solution was poured into ice–water (30 mL), and the precipitate was collected by filtration, washed with water, and dried in vacuo. Column chromatography (silica gel, 2:1 CCl_4/CH_2Cl_2) led to a first fraction which on workup was identified as 1-hydroxyanthraquinone: 14 mg (21%); mp 193–194 °C (lit.[7] mp 194–195 °C); ^{1}H NMR ($CDCl_3$) δ 12.60 (s, 1 H), 7.30–8.35 (m, 7 H); MS, *m/e* (relative intensity) 224 (100, M^+), 168 (28.5), 139 (37.5). The second fractions afforded **3** as a yellow solid: 0.051 g (68%); mp 162–164 °C; ^{1}H NMR ($CDCl_3$) δ 8.30–8.41 (m, 2 H), 7.64–7.80 (m, 3 H), 7.50–7.56 (m, 1 H), 7.08–7.12 (m, 1 H), 6.16 (q, 1 H), 1.79 (d, 3 H); ^{13}C NMR ($CDCl_3$) δ 183.12, 153.90, 151.50, 133.93, 133.88, 133.50, 132.62, 131.57, 129.74, 127.30, 124.41, 121.24, 119.39, 114.26, 87.51, 22.53; MS *m/e* (relative intensity) 249 (14.6, M^+), 234 (100), 152 (18.3), 151 (22.6), 150 (17.9). Anal. Calcd for $C_{16}H_{11}NO_2$: C, 77.10; H, 4.45; N, 5.62. Found: C, 77.35; H, 4.50; N, 5.35.

(B) Preparation from Imine 1g. The imine **1g** (0.30 g, 0.000 844 mol) was refluxed with stirring in glacial acetic acid (10 mL) for 1.5 h. The solution was cooled and added to cold water (75 mL), and the precipitate was collected by filtration, washed with water, and dried in vacuo. Chromatography as under procedure A above led to 1-hydroxyanthraquinone (0.043 g, 23%) and the 1, 3-oxazine **3** (0.141 g, 67%).

Acetone Schiff Base of 1-[(2-Aminoethyl)thio]anthracene-9,10-dione (4). To a mixture of 1-fluoroanthraquinone (**1f**; 1.00 g, 0.004 42 mol) in dioxane (25 mL) was added 2-aminoethanethiol hydrochloride (0.75 g, 0.006 64 mol). To this mixture was added a solution of sodium hydroxide (0.060 g, 0.015 mol) in ethanol–water (1:1, 6 mL) dropwise. The reaction mixture was refluxed for 1 h, cooled, and poured into water (150 mL). The precipitate was collected by filtration, washed with water, and dried in vacuo to afford a yellow-orange solid (1.17 g, 94%). Crystallization of this free base **1b** (0.400 g) from hot acetone afforded the Schiff base **4** as yellow-orange needles: (0.265 g; mp 157–159 °C; ^{1}H NMR ($CDCl_3$) δ 7.65–8.34 (m, 7 H), 3.61 (t, 2 H),

(4) Galt, R. H. B.; Loudon, J. D.; Sloan, A. D. B. *J. Chem. Soc.* **1958**, 1588.
(5) Naiki, K. *Bull. Chem. Soc. Jpn.* **1959**, *32*, 1327.
(6) Valkanas, G.; Hoppf, H. *J. Org. Chem.* **1962**, *27*, 3680.
(7) Burnett, A. R.; Thomson, R. H. *J. Chem. Soc. C* **1967**, 2100.

3.34 (t, 2 H), 2.07 (s, 3 H), 1.88 (s, 3 H); ^{13}C NMR ($CDCl_3$) δ 183.46, 183.11, 169.18, 145.09, 135.56, 134.32, 134.09, 133.63, 132.98, 132.56, 130.10, 128.97, 127.44, 126.83, 123.48, 49.84, 33.23, 29.27, 19.02; MS, *m/e* (relative intensity) 323 (1.4, M^+), 249 (10.0), 239 (14.0), 139 (14.1), 70 (100). Anal. Calcd for $C_{19}H_{17}NO_2S$: C, 70.56; H, 5.30; N, 4.33. Found: C, 70.43; H, 5.30; N, 4.21.

2,3-Dihydro-8*H*-anthra[9,1-*ef*][1,4]thiazepin-8-one (5). The crude solid **1b** (0.200 g, 0.75 mmol) was refluxed in toluene (20 mL) for 20 h. The reaction mixture was allowed to cool slightly and then introduced onto a column of silica gel. Elution with CH_2Cl and concentration yielded an orange solid as the major component (0.105 g, 56%). Crystallization from ethanol afforded **5** as orange needles: mp 180–182 °C dec; ^{1}H (NMR ($CDCl_3$) δ 7.47–8.22 (m, 7 H), 4.07–4.18 (m, 2 H), 3.80–3.90 (m, 2 H); ^{13}C NMR ($CDCl_3$) δ 183.40, 163.03, 139.62, 137.88, 137.48, 136.09, 135.41, 133.68, 131.41, 130.77, 130.24, 126.77, 126.53, 125.03, 52.14, 42.78; MS, *m/e* (relative intensity) 265 (48.3, M^+) 264 (57.5), 237 (100), 163 (52.6). Anal. Calcd for $C_{16}H_{11}NOS$: C, 72.43; H, 4.18; N, 5.28. Found: C, 72.45; H, 4.32; N, 5.22.

7*H*-Dibenzo[*de*,*h*]quinolin-7-one (6). The crude amine **1b** (0.300 g, 1.06 mmol) was refluxed in glacial acetic acid (10 mL) for 1 h. The cooled reaction mixture was poured into water (100 mL), and the precipitate was collected by filtration, washed with water, and dried in vacuo. Column chromatography (silica gel, 1:1 CCl_4/CH_2Cl_2) afforded **6** as a yellow solid (0.165 g, 66%). Crystallization from an ethanol–methanol–water mixture gave yellow needles: mp 183–185 °C (lit.[8] mp 184–186 °C; ^{1}H NMR ($CDCl_3$) δ 8.88–8.95 (m, 1 H), 8.77–8.81 (m, 1 H), 8.65–8.70 (m, 1 H), 8.40–8.47 (m, 1 H), 8.15–8.19 (m, 1 H), 7.62–7.94 (m, 4 H); ^{13}C NMR ($CDCl_3$) δ 183.91, 149.25, 144.50, 137.22, 135.59, 134.41, 133.75, 132.82, 130.90, 130.76, 130.25, 129.55, 127.95, 125.71, 123.29, 121.23; MS, *m/e* (relative intensity) 231 (100, M^+), 230 (14.5), 203 (38.1), 88 (14.6).

Acknowledgment. This research was supported by Grant CA 24543 from the National Cancer Institute.

Registry No. **1a**, 86709-68-4; **1b**, 86709-69-5; **1c**, 82-44-0; **1e**, 4465-58-1; **1f**, 569-06-2; **1g**, 86709-70-8; **1h**, 86709-71-9; **2**, 86709-72-0; **3**, 86709-73-1; **4**, 86709-74-2; **5**, 86709-75-3; **6**, 65543-67-1; 2-aminoethanol, 141-43-5; 2-(benzylidineamino)-ethanol, 770-37-6; 2-aminoethanethiol hydrochloride, 156-57-0.

(8) Bayer, O. In "Methoden der Organische Chemie (Houben-Weyl)"; Georg Thieme Verlag: Stuttgart, 1979; Band 7 (3c), p 347 and references cited therein.

Isolation of the Intermediates in a Benzoin-Type Condensation

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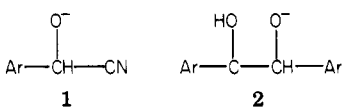
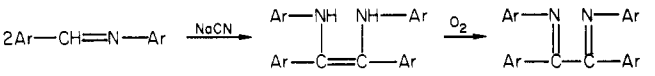
Received December 30, 1982

The mechanism of the benzoin condensation for aromatic aldehydes is well established, at least in alcoholic solution.[1] Two important cyanohydrin intermediates, **1** and **2**, exist in this reaction. Several workers have demonstrated that **1** accumulates in significant amounts during the course of the condensation, and it has been isolated both as a salt and as its conjugate acid.[1a,b] However, **2** is readily converted to a product, and, not surprizingly, a literature search has failed to turn up any report of its isolation.



1 **2**

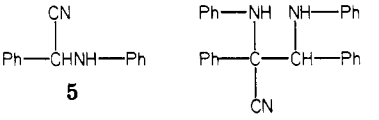
Schiff bases undergo a reaction that gives a benzoin-type product (**3**), which is rapidly oxidized to **4**.[2] The reaction



3 **4**

is assumed to proceed via the same mechanism as the benzoin condensation, but there has been no supporting evidence for this assumption.

This note reports the use of a phase-transfer agent, tetrabutylammonium chloride, in the reaction of NaCN with *N*-benzylideneaniline. When the reaction was run in a two-phased solid–liquid system with toluene as the organic solvent, **3** and **4** were obtained in moderate yield. The only other compound isolated was a decomposition product of **3**. In contrast, when performed in a two-phased methylene chloride–H_2O system, the reaction proceeded to give not only **3** (isolated as **4**) but also **5** and **6**, which



5 **6**

are the conjugate acids of the corresponding analogues of **1** and **2**. In fact, **6** is formed in nearly quantitative yield when the reaction is run in a toluene–H_2O mixture. This high yield of **6** is explained by its extreme insolubility in toluene, thus shifting all equilibria toward its formation. A reversible sequence of reactions was demonstrated by interconverting **5** and **6**, as well as converting each into **3**.[3]

Compound **5** is well-known, and it has been prepared in many ways.[4] The structure for **6** is supported by analysis and by its UV, NMR, and mass spectra. Interestingly, **6** does not exhibit a nitrile absorption in the IR. This is not unexpected, since nitriles that have an electron-withdrawing substituent α to the nitrile often have very weak or undetectable absorption in the 2000–2400-cm^{-1} region.[5] The presence of the CN functional group was demonstrated by generation and identification of CN^- from **6** in a basic solution.

The mass spectrum confirms the α-amino nitrile structure in **6**. The spectrum consists largely of two fragments at *m/e* 362 and 180. The first is due to loss of HCN from the parent molecule and the second is probably Ph–$\overset{+}{C}$≡N–Ph, resulting from fragmentation of the *m/e* 362 molecule. A molecular ion at *m/e* 389 is observable, but its intensity is very weak. The spectrum is entirely consistent with mass spectra reported for other aromatic α-amino nitriles.[6]

There has been a prior report of **6**. It was obtained as a byproduct during the reaction of carvone with **5** under

(1) (a) Lapworth, A. *J. Chem. Soc.* **1903**, *83*, 995. (b) Lapworth, A. *Ibid.* **1904**, *85*, 1206. (c) Kuebrich, J. P.; Schowen, R. L.; Wang, M.; Lupes, M. *J. Am. Chem. Soc.* **1971**, *93*, 1214.

(2) (a) Strain, H. *J. Am. Chem. Soc.* **1928**, *50*, 2218. (b) Strain H. *J. Am. Chem. Soc.* **1929**, *51*, 269. (c) Becker, H. D. *J. Org. Chem.* **1970**, *35*, 2099.

(3) Since **5** and **6** are the conjugate acids of the true intermediates, the extent of conversion of **5** and **6** into **3** depends on the basicity of the reaction medium. See Experimental Section.

(4) See, for example: (a) Everset, A. E.; McCombie, H. *J. Chem. Soc.* **1911**, *99*, 1756. (b) von Walther, R.; Hubner, R. *J. Prakt. Chem.* **1916**, *93*, 119. (c) McEwen, W. E.; Grossi, A. V.; MacDonald, R. J.; Stamegna, A. P. *J. Org. Chem.* **1980**, *45*, 1301.

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