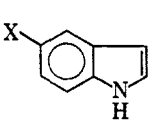
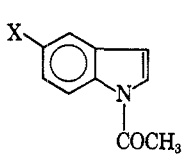
iodine monochloride should have provided 5-iodoindole, but in our hands it regenerated indole.

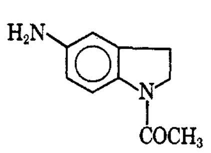
Attempted conversion of 5-aminoindole[8] to **1b** *via* a Sandmeyer reaction gave a tar from which no 5-iodoindole could be isolated. Consequently, 5-nitroindole (**1a**)[8] was acetylated to give N-acetyl-5-nitroindole (**2a**).[9] Catalytic reduction of the nitro group of **2a** using a 5% palladium on barium sulfate catalyst at 10 psig of hydrogen gave N-acetyl-5-aminoindole (**2b**) along with a small amount of N-acetyl-5-aminoindoline (**3**). (Catalytic reduction of **2a** with 5% palladium on carbon catalyst at 35 psig of hydrogen gave predominantly **3**.) Treatment of **2b** in hydrochloric acid with sodium nitrite and potassium iodide gave N-acetyl-5-iodoindole (**2c**), which on hydrolysis of the acetate function with Claisen's caustic then produced 5-indoindole (**1b**) by a relatively simple series of chemical steps in about 20% over-all yield.



1a, X = NO$_2$
b, X = I



2a, X = NO$_2$
b, X = NH$_2$
c, X = I



3

Experimental Section[10]

N-Acetyl-5-nitroindole (**2a**).—A mixture of 50 g (0.31 mole) of 5-nitroindole, 40 g of potassium acetate, and 300 ml of acetic anhydride was heated under nitrogen for 2 hr at 95°. After cooling to 5°, the mixture was filtered, the cake was washed with 30 ml of acetic anhydride, and reslurried in 200 ml of water at 55° for 5 min. Filtration and drying at 55° for 20 hr gave 53.6 g (85%) of yellow **2a**: mp 178–180° (lit.[10] mp 179.5–180°); $\lambda_{\max}^{\text{Nujol}}$ 5.85 μ; $\lambda_{\max}^{\text{MeOH}}$ 297 mμ (ϵ 9000), sh, 275 (12,750), 263 (23,300) 254 (23,000).

Anal. Calcd for $C_{10}H_8N_2O_3$: C, 58.8; H, 3.94; N, 13.7. Found: C, 59.0; H, 4.21; N, 13.5.

N-Acetyl-5-aminoindole (**2b**).—A mixture of 55 g (0.27 mole) of N-acetyl-5-nitroindole (**2a**), 5.5 g of 5% palladium on barium sulfate catalyst, and 1400 ml of absolute ethanol was hydrogenated at 10 psig for 1 hr. The mixture was filtered and the catalyst cake was washed with 50 ml of absolute ethanol. Concentration of the ethanol solution to about 200 ml and cooling gave 42 g (89%) of **2b**: mp 126–128°; $\lambda_{\max}^{\text{Nujol}}$ 2.93, 3.0, 5.94 μ; $\lambda_{\max}^{\text{MeOH}}$ 259 mμ (ϵ 20,000).

Anal. Calcd for $C_{10}H_{10}N_2O$: C, 69.0; H, 5.79; N, 16.1. Found: C, 68.8; H, 5.89; N, 16.0.

N-Acetyl-5-iodoindole (**2c**).—A mixture of 42 g (0.24 mole) of N-acetyl-5-aminoindole (**2b**), 62.4 ml of concentrated hydrochloric acid, and 124.8 ml of water at −5° was treated dropwise with a solution of 17.4 g of sodium nitrite in 36 ml of water to a positive starch–iodide test. Then a solution of 42 g of potassium iodide in 42 ml of water was added at −5°, after which the mixture was allowed to warm slowly to room temperature. The orange mixture was heated to 40°, treated with 500 ml of water and 500 ml of chloroform, and the phases were allowed to separate. The chloroform phase was washed with 500 ml each of water, 5% sodium bisulfite, 0.1 *N* hydrochloric acid, 5% sodium bicarbonate, and water in the order given. Concentration of the chloroform solution gave a 50 g residue. This was taken up in 150 ml of benzene and diluted with 150 ml of hexane to separate a tar. Further dilution with 300 ml of hexane effected complete tar removal. Concentration of the hexane solution then gave 39 g (57%) of **2c**: mp 106–107°; $\lambda_{\max}^{\text{Nujol}}$ 5.87 μ; $\lambda_{\max}^{\text{MeOH}}$ 246 mμ (ϵ 30,100).

Anal. Calcd for $C_{10}H_8INO$: C, 42.2; H, 2.83; N, 4.92. Found: C, 42.5; H, 3.00; I, 42.4; N, 4.58.

5-Iodoindole (**1b**).—A solution of 39 g (0.137 mole) of N-acetyl-5-iodoindole in 400 ml of Claisen's caustic (prepared by diluting a solution of 141 g of potassium hydroxide in 101 ml of water to 400 ml with methanol) was heated at 75–80° for 30 min. After cooling, the solution was diluted with 1000 ml of water and extracted with 1000-ml and 200 portions of benzene. The combined benzene extracts were washed with 500 ml each of water, 1% sodium bisulfite, 5% sodium bicarbonate, and water in the order given. The benzene solution was concentrated to about 40 ml, then diluted with 320 ml of hexane to effect separation of an oil. Decantation of the hexane solution from the oil and concentration gave 16 g (48%) of the typical silvery plates of **1b**: mp 99–102° (lit.[5] mp 99°); $\lambda_{\max}^{\text{Nujol}}$ 2.93, 6.40 μ; $\lambda_{\max}^{\text{MeOH}}$ sh 298 mμ (ϵ 3,210), 288 (5050), 80 (5500), 230 (44,000), 226 (46,300).

Anal. Calcd for C_8H_6IN: C, 39.5; H, 2.49; I, 52.3; N, 5.77. Found: C, 39.4; H, 2.74; I, 51.8; N, 5.70.

N-Acetyl-5-aminoindoline (**3**).—A mixture of 1.0 g (0.0049 mole) of N-acetyl-5-nitroindole (**2a**), 0.5 g of 5% palladium on carbon catalyst, and 100 ml of absolute ethanol was hydrogenated at 35 psig for 1 hr. The mixture was filtered and the catalyst cake was washed with 10 ml of absolute ethanol. Concentration of the ethanol solution gave 0.8 g (93%) of white **3**: mp 181–183°; $\lambda_{\max}^{\text{Nujol}}$ 6.10.

Anal. Calcd for $C_{10}H_{12}N_2O$: C, 68.2; H, 6.87; N, 15.9. Found: C, 67.9; H, 6.69; N, 16.1.

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[8] Available from Aldrich Chemical Co.

[9] W. E. Noland and K. R. Rush, *J. Org. Chem.*, **31**, 70 (1966).

[10] All melting points were taken with a Thomas-Hoover capillary melting point apparatus and are corrected. The infrared and ultraviolet spectra were obtained using Perkin-Elmer 621 and Cary 15 spectrophotometers. Microanalyses were determined by the Microanalytical Laboratory of the Squibb Institute for Medical Research.

The Formation and Structure of Certain Oxazolonium Compounds

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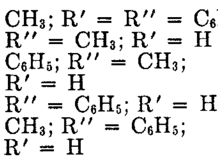
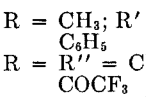
Huisgen and his co-workers[2] reported that N-substituted N-acylamino acids (**1**) readily lose a molecule of water when treated with acetic anhydride at 55° to give anhydro compounds **2**. They isolated only one compound, namely, anhydro-3-methyl-2,4-diphenyl-5-hydroxy-1,3-oxazolonium hydroxide (**2a**).





1a, R = CH_3; R′ = R″ = C_6H_5
b, R = R″ = CH_3; R′ = H
c, R = C_6H_5; R″ = CH_3; R′ = H
d, R = R″ = C_6H_5; R′ = H
e, R = CH_3; R″ = C_6H_5; R′ = H

2a, R = CH_3; R′ = R″ = C_6H_5
b, R = R″ = CH_3; R′ = $COCF_3$
c, R = C_6H_5; R″ = CH_3; R′ = $COCF_3$
d, R = R″ = C_6H_5; R′ = $COCF_3$
e, R = CH_3; R″ = C_6H_5; R′ = $COCF_3$

A short time after Huisgen's publications, we initiated a program to prepare and isolate additional derivatives of **2**, but with R′ = H, by dehydrative cyclization of

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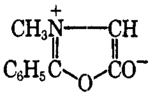
[1] To whom all inquiries should be addressed.

[2] (a) R. Huisgen, H. Gotthardt, and H. O. Bayer, *Tetrahedron Letters*, 481 (1964); (b) H. Gotthardt, R. Huisgen, and F. C. Schaeffer, *ibid.*, 487 (1964); (c) R. Huisgen, H. Gotthardt, and H. O. Bayer, *Angew. Chem. Intern. Ed. Engl.*, **3**, 135 (1964); (d) R. Huisgen, H. Gotthardt, H. O. Bayer, and F. C. Schaeffer, *ibid.*, **3**, 136 (1964).

N-substituted N-acylamino acids (**1b**–**e**). The use of acetic anhydride for the cyclodehydration failed to produce isolable mesoionic oxazolones (**2**), except in the case of N-benzoylsarcosine (**1e**).[3] Trifluoroacetic anhydride readily yielded isolable mesoionic compounds[4] but concomitantly trifluoroacetylated the unsubstituted 4 position to give **2b**–**e**.

To obtain mesoionic oxazolones unsubstituted at the 4 position, the use of dicyclohexylcarbodiimide (DCC) was then explored for the cyclodehydration of N-benzoylsarcosine (**1e**) and N-acetylsarcosine (**1b**). The expected products, anhydro-3-methyl-5-hydroxy-2-phenyl-1,3-oxazolonium hydroxide (**3**) and anhydro-2,3-dimethyl-5-hydroxy-1,3-oxazolonium hydroxide (**4**), failed to result. While elemental analyses agreed with these structures, the observed molecular weights of 380 (Rast) and 388 (thermoelectric) for the 2-phenyl-3-methyl compound and 207 (thermoelectric) for the 2,3-dimethyl compound deviated about 8% from the doubled-calculated values of 175 and 113 for **3** and **4**, respectively. These results suggested the acylated oxazolonium structure **5**.



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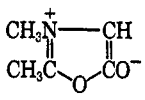

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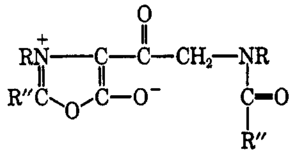
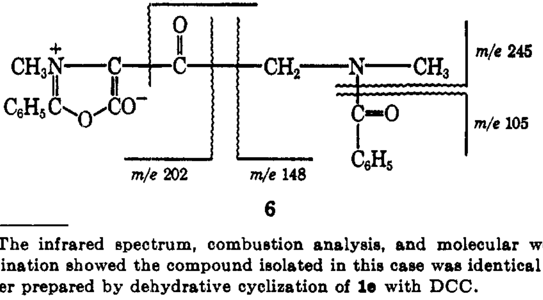


5a, R = R'' = CH_3
b, R = CH_3; R'' = C_6H_5

The infrared spectra (potassium bromide disk) of **2b**–**e** showed an intense band at 1625 cm^{-1}, attributed to the iminium grouping in the ring, and a doublet which was most pronounced in the dimethyl compound **2b** at 1800 and 1785 cm^{-1}, attributed to the trifluoroacetylcarbonyl and ring carbonyl, respectively.[5] Compounds **5a** and **b** also showed an intense band at 1620 cm^{-1} and the doublet in the dimethyl compound (**5a**) at 1775 and 1750 cm^{-1} which could be attributed to a ketonic carbonyl and a ring carbonyl, respectively.

A mass spectral analysis of **5b** gave the parent radical ion at *m/e* 350 (mol wt 350). The most intense peak at *m/e* 105 was attributed to an acylium fragment formed by cleavage of the amide bond as indicated in structure **6**. A peak at *m/e* 245 results from a cleavage at this point. The peak at *m/e* 77, considering the magnitude of its relative abundance, results from further fragmentation of the acylium ion with loss of carbon monoxide. Two peaks at *m/e* 202 and 148 indicate fragmentation of the bond to the oxazolonium carbonyl. An additional cleavage of the bond to the carbonyl was indicated by the peak at *m/e* 175 (176-H). Some decomposition of the aromatic oxazolonium ring was apparent from the *m/e* 217 peak resulting from loss of carbon monoxide from ion **7**.



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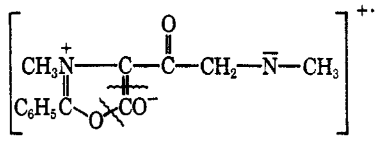
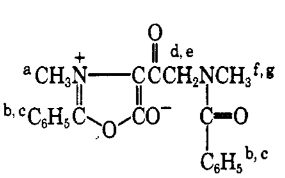


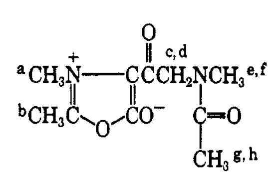
7

The results of a nuclear magnetic resonance (nmr) spectral analysis (deuteriochloroform, 30°) on **5a** and **b** are summarized (in parts per million). With both compounds, the peak separations observed at 30° for the methylene and side-chain N-methyl groups, as well as for the acyl methyl group of **5a**, were a result of formation of a partial double bond (possible resonance hybrid) producing restricted rotation about the $-\overset{|}{N}-\overset{|}{C}{=}O$ bond. The methylene group absorbance of **5b** could be expected to appear at lower field when the methylene group was *cis* with the oxygen nucleus than when *cis* with the phenyl ring. Also, with two different substituents on the amide nitrogen atom, the two methylene protons could never occupy identical environments in either **5a** or **b**. The N-methyl signal for **5b** is separated into two absorptions approximately 4 cps apart while for **5a** a peak separation of about 10 cps was observed for each methyl group in the side chain. There is no evidence to indicate that these peak separations are due to spin–spin coupling. A spectrum of **5a** at 94° (dimethyl sulfoxide-d_6) showed that the methylene, N-methyl, and acyl methyl group absorbances coalesced into three bands each occupying a chemical shift intermediate to the peaks observed at 30°.



5b

a,	4.15	e,	4.76
b,	7.39	f,	3.04
c,	7.67	g,	3.10
d,	4.52		



5a

a,	3.92	e,	2.90
b,	2.55	f,	3.02
c,	4.42	g,	2.11
d,	4.47	h,	1.95

Experimental Section

All melting points were determined on a Mel-Temp melting point apparatus and are uncorrected. All combustion analyses were performed by Schwarzkopf Microanalytical Laboratories, Woodside, N. Y. Ultraviolet absorption spectra were determined with a Bausch and Lomb Spectronic 505 spectrophotometer, all samples being dissolved in chloroform. Infrared spectra were measured on a Perkin-Elmer Model 137 using the potassium bromide pellet technique. Mass spectra were measured with an Hitachi Perkin-Elmer Model RMU-6D spectrometer by Morgan

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[3] The infrared spectrum, combustion analysis, and molecular weight determination showed the compound isolated in this case was identical with **5b**, later prepared by dehydrative cyclization of **1e** with DCC.

[4] While we were successfully preparing the trifluoroacetylated compounds, G. Singh and S. Singh [*Tetrahedron Letters*, 3789 (1964)] communicated the synthesis of one of our compounds. In view of the fact that the experimental details were omitted by these workers, our syntheses and spectral results are included here.

[5] W. D. Burrows, *J. Org. Chem.*, **31**, 3435 (1966).

Schaffer, Quebec, Canada, ionizing energy 70 ev. Nmr spectra were obtained on a Varian A60A spectrometer (Sadtler Research Corp.) and data are expressed in parts per million downfield from an internal tetramethylsilane standard.

Materials.—N-Methylglycine (sarcosine) (mp 210–211°) was obtained from Nutritional Biochemicals Co., Cleveland, Ohio. N-Phenylglycine (mp 125–127°) and N-acetyl-N-phenylglycine (mp 190–192°) were obtained from Eastman Organic Chemicals, Rochester, N. Y. N-Benzoyl-N-methylglycine (N-benzoylsarcosine) was prepared using O'Brien and Niemann's procedure,[6] mp 104–105°. N-Acetyl-N-methylglycine (N-acetylsarcosine) was prepared according to Southwick's procedure[7] (mp 136–137.5°) and N-benzoyl-N-phenylglycine was obtained by Rebuffat's method.[8]

Anhydro-2,3-dimethyl-4-trifluoroacetyl-5-hydroxy-1,3-oxazolonium Hydroxide (2b).—N-Acetylsarcosine (1 g, 0.007 mole) was added to 10 ml of trifluoroacetic anhydride and the mixture was stirred for 1.5 hr at room temperature. The resulting solution was then poured into 50 ml of dry ether, thereby precipitating 1.21 g (83%) of white crystals, mp 128–131°. Several recrystallizations from benzene–petroleum ether (bp 30–60) gave 0.96 g (65%) of white crystals, mp 133–135°. The ultraviolet spectrum showed $\lambda_{\max}$ at 310 mμ (ϵ 1.83 × 10^5) and 263 mμ (ϵ 7.5 × 10^4).

Anal. Calcd for $C_7H_6F_3NO_3$; C, 40.20; H, 2.89; F, 27.25; N, 6.70. Found: C, 40.36; H, 3.09; F, 27.51; N, 6.54.

Anhydro-3-phenyl-4-trifluoroacetyl-5-hydroxy-2-methyl-1,3-oxazolonium hydroxide (2c) was prepared as above using 2 g (0.010 mole) of N-acetyl-N-phenylglycine in 20 ml of trifluoroacetic anhydride. The crude yield of crystals (mp 202–205°) was 2.4 g (88.4%). Several recrystallizations from a minimum volume of boiling ethyl acetate gave 0.6 g (22%) of crystals, mp 211–212°. The ultraviolet spectrum showed $\lambda_{\max}$ at 264 mμ (ϵ 6.47 × 10^3) and 312 mμ (ϵ 1.43 × 10^4).

Anal. Calcd for $C_{12}H_8F_3NO_3$: C, 53.14; H, 2.97; F, 21.02; N, 5.17. Found: C, 53.15; H, 3.01; F, 21.05; N, 5.18.

Anhydro-2,3-diphenyl-4-trifluoroacetyl-5-hydroxy-1,3-oxazolonium hydroxide (2d) was prepared as was **2b** whereby 1.0 g (0.0039 mole) of crude N-benzoyl-N-phenylglycine was added to 10 ml of trifluoroacetic anhydride. The crude yield of bright yellow crystals which precipitated was 1.19 g (91%), mp 185–190°. Several recrystallizations from benzene–petroleum ether gave 1.10 g (84%) of yellow crystals, mp 193–195° (lit.[4] value 195°). The ultraviolet spectrum showed $\lambda_{\max}$ at 367 (ϵ 1.86 × 10^4), 276 (1.26 × 10^4), and 246 mμ (1.36 × 10^4).

Anal. Calcd for $C_{17}H_{10}F_3NO_3$: C, 61.27; H, 3.02; F, 17.10; N, 4.20. Found: C, 61.17; H, 3.32; F, 16.84; N, 4.23.

Anhydro-3-methyl-4-trifluoroacetyl-5-hydroxy-2-phenyl-1,3-oxazolonium hydroxide (2e) was prepared as was **2b** using 1.0 g (0.005 mole) of N-benzoylsarcosine in 10 ml of trifluoroacetic anhydride. The crude yield of yellow crystals was 1.30 g (96%), mp 161–164°. Several recrystallizations from benzene–petroleum ether gave 0.90 g (66%) of yellow crystals, mp 161.5–163°. The ultraviolet spectrum showed $\lambda_{\max}$ at 347 (ϵ 2.29 × 10^4), 276 (1.24 × 10^4), and 243 mμ (1.14 × 10^4).

Anal. Calcd for $C_{12}H_8F_3NO_3$: C, 53.15; H, 2.97; F, 21.02; N, 5.17; mol wt, 271.2. Found: C, 53.10; H, 3.03; F, 20.77; N, 5.29; mol wt, 260 (Rast).

Anhydro-2,3-dimethyl-4-(N-acetylsarcosyl)-5-hydroxy-1,3-oxazolonium Hydroxide (5a).—To 4.24 g (0.034 mole) of N-acetylsarcosine in 50 ml of nitromethane was added 7.00 g (0.034 mole) of DCC dissolved in 50 ml of nitromethane. After 2 hr of stirring the solution was filtered and the solid remaining behind was rinsed with nitromethane to give 7.13 g (94%) of dicyclohexylurea, mp 232.5–234.5° (lit.[9] value 234°). The filtrate was evaporated *in vacuo* to give an oil. Ethyl acetate (15 ml) was added and after a few days' refrigeration, off-white crystals deposited. They were filtered to give 2.15 g (56%), mp 134–137°. After two recrystallizations from a minimum volume of boiling ethyl acetate, 1.00 g (26%) was recovered, mp 137–138°. The ultraviolet spectrum showed $\lambda_{\max}$ at 300 mμ (ϵ 1.35 × 10^4) and 260 mμ (ϵ 9.78 × 10^3).

Anal. Calcd for $C_{10}H_{14}N_2O_4$: C, 53.08; H, 6.24; N, 12.38; mol wt, 226. Found: C, 53.11; H, 6.27; N, 12.21; mol wt, 207 (thermoelectric).

Anhydro-3-methyl-4-(N-benzoylsarcosyl)-5-hydroxy-2-phenyl-1,3-oxazolonium Hydroxide (5b). A. Using DCC.—To 6.55 g (0.034 mole) of N-benzoylsarcosine in 50 ml of nitromethane was added 7.00 g (0.034 mole) of DCC dissolved in 50 ml of nitromethane. After 2 hr of stirring the solution was filtered and the solid remaining behind was rinsed with nitromethane to give 6.90 g (91%) of DCU, mp 232–235°. The filtrate was evaporated *in vacuo* to give a dark red oil. Ethyl acetate (25 ml) was added, the solution was boiled, and upon cooling crystals were deposited, yield 2.18 g (37%), mp 169–172°. The solid was recrystallized from a minimum volume of boiling ethyl acetate to give 1.32 g (22%) of yellow crystals, mp 175–176°. The ultraviolet spectrum showed $\lambda_{\max}$ at 352 (ϵ 1.61 × 10^4), 268 (1.24 × 10^4), and 244 mμ (1.24 × 10^4).

Anal. Calcd for $C_{20}H_{18}N_2O_4$: C, 68.55; H, 5.18; N, 8.00; mol wt, 350. Found: C, 68.43; H, 5.25; N, 8.26; mol wt, 380 (Rast), 388 (thermoelectric), 350 (mass spectrometry).

B. Using Acetic Anhydride.—To 5.0 g (0.026 mole) of N-benzoylsarcosine in a 100-ml round-bottom flask was added 50 ml of acetic anhydride and the mixture was stirred at 55° for 1 hr in an oil bath. After overnight refrigeration, the excess acetic anhdride was evaporated *in vacuo*. To remove excess anhydride residue, the oil was washed several times with dry ether. The resulting yellow solid was dissolved in boiling ethyl acetate and the solution was evaporated on a steam bath to a volume of 50 ml. After several days' refrigeration, a solid was deposited, yield 2.15 g (44%), mp 165–169°. Two recrystallizations from a minimum volume of boiling ethyl acetate gave 1.15 g (39%) of yellow crystals, mp 175.5–177°.

A mixture melting point with the anhydro-3-methyl-4-(N-benzoylsarcosyl)-5-hydroxy-2-phenyl-1,3-oxazolonium hydroxide (**5b**) made using DCC showed no depression.

The ultraviolet (as well as infrared) spectrum showed the same absorptions as compound (**5b**).

Anal. Calcd for $C_{20}H_{18}N_2O_4$: C, 68.55; H, 5.18; N, 8.00; mol wt, 350. Found: C, 68.88; H, 5.10; N, 8.14; mol wt, 375 (thermoelectric).

Registry No.—**2b**, 14119-97-2; **2c**, 14119-98-3; **2d**, 14172-82-8; **2e**, 14119-99-4; **5a**, 14120-00-4; **5b**, 14120-01-5.

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[6] J. O'Brien and C. Niemann, *J. Am. Chem. Soc.*, **79**, 84 (1957).

[7] P. Southwick, H. Diamond, and R. Stansfield, *ibid.*, **78**, 1609 (1956).

[8] G. Rebuffat, *Gazz. Chim. Ital.*, **17**, 232 (1887).

[9] Noel F. Albertson, *Org. Reactions*, **12**, 212 (1962).

Synthesis and Properties of Fluorine-Containing Heterocyclic Compounds. IV. An N,N'-Unsubstituted Imidazolidine[1]

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Received April 20, 1967

The reaction of ethyl acetoacetate with *o*-phenylenediamine may yield several products depending upon conditions.[3,4] In contrast, only diethyl (3,3'-ethylenedimino)dicrotonate was isolated from the reaction with 1,2-ethylenediamine (I);[5] no product was isolated from the reaction of I and ethyl trifluoroacetoacetate.[6a] In continuation of our previous studies,[7] we have now re-

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[1] Part III in this series: A. S. Dey and M. M. Joullié, *J. Org. Chem.*, **30**, 3237 (1965).

[2] To whom all correspondence should be addressed.

[3] J. Davoll, *J. Chem. Soc.*, 308 (1960).

[4] A. Rossi, A. Hunger, J. Kebrle, and K. Hoffmann, *Helv. Chim. Acta*, **43**, 1298 (1960).

[5] C. M. Hofmann and S. R. Safir, *J. Org. Chem.*, **27**, 3565 (1962).

[6] (a) A. E. Martell, R. L. Belford, and M. Calvin, *J. Inorg. Nucl. Chem.*, **5**, 170 (1958); (b) G. O. Dudek and R. H. Holm, *J. Am. Chem. Soc.*, **83**, 2099 (1961).

[7] (a) M. M. Joullié, S. Násfay, and L. Rypstat, *J. Org. Chem.*, **21**, 1358 (1956); (b) F. B. Wigton and M. M. Joullié, *J. Am. Chem. Soc.*, **81**, 5212 (1959); (c) A. S. Dey and M. M. Joullié, *J. Heterocyclic Chem.*, **2**, 113, 120 (1965); (d) D. K. Wald and M. M. Joullié, *J. Org. Chem.*, **31**, 3369 (1966).