

between fluorene on the one hand and toluene on the other.

Since anthracene is insoluble in carbon tetrachloride a series of experiments was made in carbon disulfide. Only 0.005 mole of the hydrocarbon is dissolved in 50.00 ml. of carbon disulfide, and the previous experimental procedure repeated at 26°. To make the iodine effects comparable, iodine-carbon disulfide solutions were prepared containing the same amount of iodine as in Table I. Only 0.005 mole of fluorene in 50.00 ml. of solvent was used also.

TABLE II

CARRIER EFFECT OF IODINE ON DARK ROOM BROMINATION IN CS<sub>2</sub>, 26°

I <sub>2</sub> concn. =	0.290		0.029		0.000	
	Time, min.	%	Time, min.	%	Time, min.	%
Fluorene	10	31 ± 2	10	5 ± 1	60	6 ± 2
Anthracene	1	76 ± 1	..	...	1	60 ± 1

Anthracene is so rapidly brominated even in the absence of iodine that it is possible to determine only from Table II that the effect is positive.

In a final series of brominations when the experiments were run overnight in a darkened laboratory at room temperature with and without iodine in carbon tetrachloride solution, the results were as follows: In the absence of iodine

there was no perceptible bromination with toluene, dibenzyl, fluorenone, 9,10-anthraquinone and 2-methyl-9,10-anthraquinone; in the presence of iodine toluene showed a bromination of 55, dibenzyl, 51, and fluorenone, 5%, respectively, while the others were unaffected.

The authors are grateful to Dr. E. Emmet Reid for his active interest in this research, and to the Office of Naval Research for a grant.

### Summary

1. An approximate method has been developed for measuring the rate of bromination of condensed ring compounds, using standard sodium sulfite solution.

2. Rates of dark room bromination, both with and without iodine as carrier, have been measured for the following compounds: fluorene, fluorenone, naphthalene, anthracene, phenanthrene, dibenzyl, diphenyl, diphenylmethane, triphenylmethane and toluene. 9,10-Anthraquinone and 2-methyl-9,10-anthraquinone are not brominated in overnight experiments at room temperature, either with or without iodine.

3. Fluorene, phenanthrene, anthracene and naphthalene all show an enhanced effect of iodine as carrier.

GREENVILLE, SOUTH CAROLINA

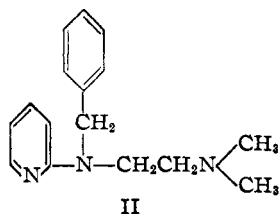
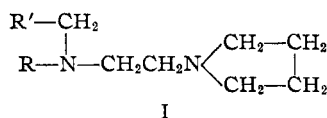
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## Histamine Antagonists. VI. Pyrrolidylethylamine Derivatives

BY EDWARD H. LINCOLN, R. V. HEINZELMANN AND JAMES H. HUNTER

In the course of an extensive investigation of histamine antagonists being conducted in this Laboratory<sup>1-5</sup> a number of tertiary pyrrolidylethylamine derivatives of general structure I have been prepared and their antihistaminic activity determined.



(1) Wright, Koloff and Hunter, *This Journal*, **70**, 3098 (1948).

(2) Reid, Wright, Koloff and Hunter, *ibid.*, **70**, 3100 (1948).

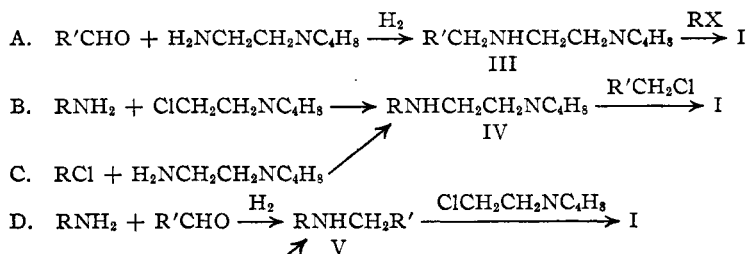
(3) Reitsema and Hunter, *ibid.*, **70**, 4009 (1948).

(4) Wright, *ibid.*, **71**, 1028 (1949).

(5) Wright, *ibid.*, **71**, 2035 (1949).

The groups R and R' were varied widely, as indicated in Table II. In general, the order of activity of these pyrrolidylethylamine compounds is low<sup>6</sup>; the most active members, N-(2-pyridyl)-N-(p-methoxybenzyl)-β-(1-pyrrolidyl)-ethylamine and N-(2-pyridyl)-N-(5-chloro-2-thenyl)-β-(1-pyrrolidyl)-ethylamine, had an effectiveness equal to approximately one-fourth that of II.

The tertiary amines were prepared via the corresponding secondary amino compounds by one of the following procedures.



(6) The assays were carried out under the direction of Dr. Milton J. Vander Brook of our Pharmacology Department.

TABLE I

SECONDARY AMINES											
R	R <sup>1</sup> CH <sub>2</sub>	Condensing agent	Pro- cedure	°C.	B. p. Mm.	Yield, %	Salt	M. p., °C.	Nitrogen, <sup>a</sup> % Calcd.	% Found	
Type III, R <sup>1</sup> CH <sub>2</sub> NHCH <sub>2</sub> CH <sub>2</sub> N						$\begin{matrix} \diagup \text{CH}_2-\text{CH}_2 \\ \diagdown \text{CH}_2-\text{CH}_2 \end{matrix}$					
	2-Furfuryl		A	94-97	0.30	60	2 HCl	212	10.49	10.80	
	2-Thenyl		A	92-102	.40	74	2 HCl	209-210	9.89	9.99	
	<i>p</i> -Methoxybenzyl		A	135-138	.30	62	2 HCl	208-208.5	9.12	9.18	
Type IV, RNHCH <sub>2</sub> CH <sub>2</sub> N						$\begin{matrix} \diagup \text{CH}_2-\text{CH}_2 \\ \diagdown \text{CH}_2-\text{CH}_2 \end{matrix}$					
2-Pyridyl	NaNH <sub>2</sub> (excess amine)	B <sup>b</sup> (C)	123-124	0.22	55(74)	HCl	148-149	18.45	18.11		
2-Pyrimidyl	NaNH <sub>2</sub>	B	123-125	.55	32	Picrat <sup>c</sup>	155.5-157	23.27	23.13		
Phenyl	Na <sub>2</sub> CO <sub>3</sub>	B <sup>c</sup>	128-129	.15	74	2 HCl	173-175	10.64	10.65		
Cyclohexyl		<sup>d</sup>	93-95	.30	66	2 HCl	210	10.41	10.35		
Type V, RNHCH <sub>2</sub> R <sup>1</sup>											
2-Pyridyl	2-Furfuryl		D			14	HCl	164-165	13.30	12.51 <sup>e</sup>	
2-Pyrimidyl	<i>p</i> -Methoxybenzyl		D <sup>f</sup>	M. p. 94-97		53	HCl	161-162	16.69	16.72	
2-Thenyl	2-Thenyl	Excess amine	E	171-181 <sup>g</sup>	12.0	55	HCl	240-241 <sup>h</sup> (dec.)	5.70	5.30	

<sup>a</sup> Carbon and hydrogen analyses also satisfactory but omitted for brevity. <sup>b</sup> Procedure used by Whitmore, *et al.*, THIS JOURNAL, 67, 393 (1945), and Huttner, *et al.*, *ibid.*, 68, 1999 (1946), for the dimethyl analog. <sup>c</sup> Procedure used by Leonard and Solmssen, *ibid.*, 70, 2064 (1948), for the dimethyl analog. <sup>d</sup> Prepared by reductive amination of cyclohexanone with pyrrolidylethylamine. <sup>e</sup> Calcd.: C, 57.01; H, 5.26. Found: C, 57.02; H, 5.24. <sup>f</sup> Tschitschibabin and Knunjanz, *Ber.*, 64, 2839 (1931). <sup>g</sup> Hartough, THIS JOURNAL, 68, 1389 (1946), b. p. 150-152 at 10 mm. See ref. 12. <sup>h</sup> Hartough, *ibid.*, 70, 4018 (1948), m. p. 250-251°; our compd. had the following analysis. Calcd.: C, 48.86; H, 4.92; Cl, 14.43; N, 5.70. Found: C, 48.67; H, 4.90; Cl, 14.21; N, 5.30.

TABLE II

TERTIARY AMINES, R-NCH <sub>2</sub> CH <sub>2</sub> N $\begin{matrix} \diagup \text{CH}_2-\text{CH}_2 \\ \diagdown \text{CH}_2-\text{CH}_2 \end{matrix}$											
R	R <sup>1</sup> CH <sub>2</sub>	Type secondary amine used	Condensing agent	°C.	B. p. Mm.	Yield, %	Salt	M. p., °C.	Nitrogen, <sup>a</sup> %		Activity <sup>b</sup>
2-Pyridyl	<i>p</i> -Methoxybenzyl <sup>c</sup>	IV	NaNH <sub>2</sub>	193-198	0.2	76	2 HCl	169.5-171.5	10.93	10.94	1/4
2-Pyridyl	2-Furfuryl	III	Na <sub>2</sub> CO <sub>3</sub>	149-153	.25	29	Citrate	133-138			1/10
							Dipicrate	152-154	17.28	17.04	
2-Pyridyl	2-Thenyl	III	Na <sub>2</sub> CO <sub>3</sub>	155-165	.5	29	Citrate	111-113	8.76	8.72	1/10
2-Pyridyl	5-Chloro-2-thenyl	IV	NaNH <sub>2</sub>	168-172	.1	56	Citrate	117-118	8.18	8.22	1/6
2-Pyrimidyl	<i>p</i> -Methoxybenzyl	V	NaNH <sub>2</sub>	203-206	.6	54	HCl	135-136	16.06	15.81	1/30-1/60
2-Pyrimidyl	5-Chloro-2-thenyl	IV	NaNH <sub>2</sub>	180-181.5	.23	18	Citrate	125-126	10.88	10.79	1/10-1/30
2-Quinoliny	2-Furfuryl	III	Na <sub>2</sub> CO <sub>3</sub>	188	1.6	38	Citrate	135-136	8.18	8.15	Sl. >1/100
2-Quinoliny	2-Thenyl	III	Na <sub>2</sub> CO <sub>3</sub>	155-160	0.2	22	HCl	246.5-247.5	11.21	11.15	<1/100
Phenyl	2-Thenyl	IV	Na <sub>2</sub> CO <sub>3</sub>	198-199	1.3	51	Citrate	126-130	5.86	6.11	1/10-1/15
Cyclohexyl	2-Thenyl	IV	K <sub>2</sub> CO <sub>3</sub>	153-154	0.2	48	Citrate	95-97	5.78	5.71	<1/100
2-Thenyl	2-Thenyl	III	Na <sub>2</sub> CO <sub>3</sub>	167-170	0.25	66	Citrate	97-100	5.62	5.64	1/100
							Dipicrate	176-177			
2-Pyridyl	Pyrrolidylethyl	<sup>d</sup>	NaNH <sub>2</sub>	178-179	0.45	28	Citrate	150-151	11.66	11.48	<1/100

<sup>a</sup> Carbon and hydrogen analyses also satisfactory but omitted for brevity. <sup>b</sup> Compared with II which is assigned an activity of 1.0. These comparisons were carried out on isolated small intestine of the guinea pig, ref. 6. <sup>c</sup> *p*-Methoxybenzyl chloride prepared by procedure of Shriner and Hull, *J. Org. Chem.*, 10, 228 (1945). <sup>d</sup> Tertiary amine formed as a by-product in the preparation of N-(2-pyridyl)-β-(1-pyrrolidyl)-ethylamine by procedure B.

In general, where applicable, Procedure A appeared to be the most suitable with respect to yields and ease of purification. Secondary amines containing the furan ring tended to discolor in the presence of mineral acid but in all cases pure white hydrochlorides were obtained in good yield after recrystallization. The tertiary amines were obtained by alkylation of the corresponding secondary amino compounds in the presence of sodamide in benzene or xylene or in the presence of sodium carbonate without solvent. In many

cases the latter condensing agent caused the formation of large amounts of tar, almost insoluble in either the water or organic layer. For the most part sodamide gave better yields and products more amenable to purification. The bases, with but few exceptions, were characterized as their citrates, which were easily purified and were relatively non-hygroscopic in contrast to many of the corresponding hydrochlorides. Those amines containing the furan ring were not found to be as unstable in the presence of mineral acid

as might be expected from the reported instability of similar compounds in the ethylenediamine series.<sup>7</sup>

### Experimental<sup>8</sup>

#### Secondary Amines

**Procedure A. N-( $\beta$ -Methoxybenzyl)- $\beta$ -(1-pyrrolidyl)-ethylamine.**—Thirteen and six-tenths grams (0.10 mole) of anisaldehyde and 11.4 g. (0.10 mole) of pyrrolidyl-ethylamine<sup>9</sup> were dissolved in 50 ml. of ethanol with cooling. The solution was hydrogenated in the presence of platinum oxide at an initial hydrogen pressure of 50 lb. The catalyst was removed by filtration, the filtrate concentrated and the residue distilled. There was obtained 14.5 g. (62%) of a colorless oil, b. p. 135–138° at 0.3 mm., which readily gave a white crystalline dihydrochloride, m. p. 208–208.5°.

*Anal.* Calcd. for  $C_{14}H_{24}Cl_2N_2O$ : C, 54.72; H, 7.87; N, 9.12. Found: C, 54.44; H, 7.54; N, 9.18.

**Procedure B. N-(2-Pyridyl)- $\beta$ -(1-pyrrolidyl)-ethylamine Hydrochloride.**—Nine and four-tenths grams (0.10 mole) of 2-aminopyridine and 4.1 g. (5% excess) of sodamide in 100 ml. of dry xylene were heated under reflux with stirring for two and one-half hours. A solution of 14 g. (5% excess) of pyrrolidylethyl chloride<sup>1</sup> in 25 ml. of xylene was added dropwise and the resulting mixture stirred under reflux for an additional seventeen to twenty hours. After cooling, the deep brown solution was filtered from all solid material, concentrated on the steam-bath at reduced pressure and the residual oil distilled yielding the desired amine (55%) which boiled at 123–124° at 0.2 mm. The hydrochloride was formed in a mixture of alcoholic hydrogen chloride and ethyl acetate; m. p. 148–149°.

*Anal.* Calcd. for  $C_{11}H_{13}ClN_3$ : C, 58.01; H, 7.96; N, 18.45. Found: C, 58.33; H, 7.94; N, 18.11.

**Procedure C. N-(2-Pyridyl)- $\beta$ -(1-pyrrolidyl)-ethylamine.**—A stirred mixture of 22.8 g. (0.20 mole) of pyrrolidylethylamine<sup>9</sup> and 15.8 g. (0.10 mole) of 2-bromopyridine was heated at 120° for nineteen hours. Water was added to the cooled mixture and the oil extracted with ether. After drying, the solvent was removed and the amine distilled, b. p. 148° at 7 mm. The yield was 8.5 g. (74% based on the 2-bromopyridine recovered (6.0 g.)).

**Procedure D. N-(2-Furfuryl)-2-aminopyridine.**—Nine-ten and two-tenths grams (0.20 mole) of freshly distilled furfural and 18.8 g. (0.20 mole) of 2-aminopyridine were refluxed gently in dry benzene for thirty minutes. The benzene was removed *in vacuo*, the gummy residue dissolved in ether which on evaporation yielded a semi-crystalline solid. The latter was triturated under petroleum ether, giving 22 g. (64%) of the yellow Schiff base melting at 87.5°. The latter was hydrogenated in ethanol in the presence of Raney nickel<sup>10</sup> under an initial hydrogen pressure of 50 lb. The free base was converted to the hydrochloride which, after recrystallization from an ethanol-ethyl acetate mixture, melted at 164–165°; yield from the Schiff base, 22%.

*Anal.* Calcd. for  $C_{10}H_{11}ClN_2O$ : C, 57.01; H, 5.26. Found: C, 57.02; H, 5.24.

**Procedure E. Di-(2-thenyl)-amine Hydrochloride.**—Forty-five and two-tenths grams (0.40 mole) of 2-thenylamine<sup>11</sup> was heated to 100° with stirring and 26.5 g. (0.20

mole) of 2-thenyl chloride<sup>11a</sup> was added dropwise over a period of one hour. After heating for an additional four hours the mixture was cooled and 10 g. of sodium hydroxide in 50 ml. of water was added. The resulting oil was extracted with ether, the solvent removed, and the base distilled. There was obtained 23 g. (55%) of the secondary amine, b. p. 171–181° at 12 mm.<sup>12</sup>

The hydrochloride [m. p. 240–241° (dec.)] was prepared in alcoholic hydrogen chloride and ethyl acetate.

*Anal.* Calcd. for  $C_{16}H_{18}ClNS_2$ : C, 48.86; H, 4.92; Cl, 14.43; N, 5.70. Found: C, 48.67; H, 4.90; Cl, 14.21; N, 5.30.

**N-Cyclohexyl- $\beta$ -(1-pyrrolidyl)-ethylamine.**—To 19.6 g. (0.20 mole) of cyclohexanone and 22.8 g. (0.20 mole) of pyrrolidylethylamine<sup>9</sup> in 100 ml. of ethanol was added 0.1 g. of Adams platinum oxide catalyst. Hydrogenation was carried out under an initial hydrogen pressure of 50 lb. and was complete in seven hours.<sup>13</sup> After filtration, the solvent was removed and the residue distilled. A colorless oil was obtained which weighed 26 g. (66%) and boiled at 93–95° at 0.3 mm.

The dihydrochloride, obtained from a mixture of alcoholic hydrogen chloride and ethyl acetate, melted at 210°.

*Anal.* Calcd. for  $C_{12}H_{22}Cl_2N_2$ : C, 53.52; H, 9.73; N, 10.41. Found: C, 53.57; H, 9.73; N, 10.35.

#### Tertiary Amines

**N-Phenyl-N-(2-thenyl)- $\beta$ -(1-pyrrolidyl)-ethylamine.**—To a stirred mixture of 6.6 g. (0.035 mole) of N-phenyl- $\beta$ -(1-pyrrolidyl)-ethylamine and 3.7 g. of anhydrous sodium carbonate at 100° was added dropwise 4.5 g. (0.035 mole) of 2-thenyl chloride. After stirring and heating at 160° for two hours,<sup>14</sup> 50 ml. of water was added and the insoluble basic material extracted with ether. The ethereal solution was dried, the solvent removed and the residual oil distilled to give 5.1 g. (51%) of the base, b. p. 198–199° at 1.3 mm. Addition of a molecular equivalent of citric acid in ethanol to an ethyl acetate solution of the base gave 8.0 g. (93%) of a white crystalline citrate, m. p. 126–130°.

*Anal.* Calcd. for  $C_{23}H_{30}N_2O_7S$ : C, 57.72; H, 6.32; N, 5.86. Found: C, 57.62; H, 6.25; N, 6.11.

**N-(2-Pyridyl)-N-(5-chloro-2-thenyl)- $\beta$ -(1-pyrrolidyl)-ethylamine.**—Ten and one-half grams (0.055 mole) of N-(2-pyridyl)-pyrrolidylethylamine and 2.2 g. (5% excess) of sodamide in 75 ml. of dry benzene were heated under reflux for one hour with vigorous stirring. After cooling, 9.2 g. (0.055 mole) of 5-chloro-2-thenyl chloride<sup>15</sup> in 25 ml. of dry benzene was added dropwise and the mixture allowed to reflux for an additional three hours. The basic material was extracted with dilute hydrochloric acid and isolated by the addition of alkali and extraction with ether. The ether layer was dried, the solvent removed and the oil distilled, giving 8.5 g. (56%)<sup>16</sup> of a yellow oil, b. p. 168–172° at 0.1 mm. The tertiary amine gave a white crystalline citrate from ethanol and ethyl acetate; m. p. 117–118°, yield 90%.

*Anal.* Calcd. for  $C_{22}H_{23}ClN_3O_7S$ : C, 51.41; H, 5.49; N, 8.18. Found: C, 51.32; H, 5.35; N, 8.22.

#### Summary

Ten secondary and twelve tertiary pyrrolidyl-ethylamines have been prepared and the latter

(7) Vaughan and Anderson, *THIS JOURNAL*, **70**, 2607 (1948). Results similar to ours were obtained by Kyrides and Zienty, *ibid.*, **71**, 1122 (1949).

(8) Melting points are uncorrected. Microanalyses by Mr. Harold Emerson and staff of our Microanalytical Laboratory.

(9) Ridi [*Gazz. chim. ital.*, **71**, 462 (1941); *C. A.*, **37**, 1123 (1943)] reported m. p. 85°.

(10) Pavlic and Adkins, *THIS JOURNAL*, **68**, 1471 (1946).

(11) (a) Blicke and Burckhalter, *ibid.*, **64**, 477 (1942); (b) since this work was completed an improved preparation of 2-thenylamine has been reported [Hartough and Meisel, *ibid.*, **70**, 4018 (1948)].

(12) The following boiling points are reported for this compound: 150–152° at 10 mm. [Hartough, Lukasiewicz and Murray, *ibid.*, **68**, 1389 (1946); 134–135° at 3 mm. [Hartough, Lukasiewicz and Murray, *ibid.*, **70**, 1146 (1948); 162–165° at 5 mm. [Hartough and Meisel, *ibid.*, **70**, 4018 (1948)].

(13) Raney nickel<sup>10</sup> gave equivalent yields but required a longer time.

(14) Reaction time was dependent on the activity of the halide used; thus, 2-bromopyridine required at least twenty hours.

(15) Clapp, *et al.*, *THIS JOURNAL*, **69**, 1549 (1947).

(16) Clapp, *et al.*,<sup>15</sup> using a similar procedure, reported a 62% yield of the dimethyl analog.

assayed for antihistaminic activity. On the basis of preliminary pharmacological data, the most active members of the series possessed an activity

equal to one-fourth that of N,N-dimethyl-N'-benzyl-N'-( $\alpha$ -pyridyl)-ethylenediamine.

KALAMAZOO, MICHIGAN

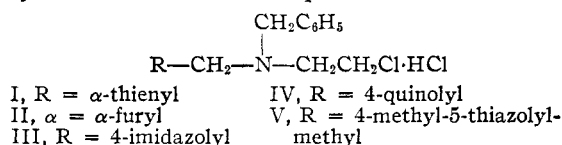
RECEIVED MARCH 18, 1949

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, UNIVERSITY OF NOTRE DAME]

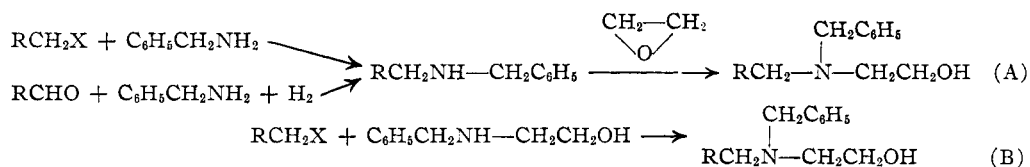
## The Preparation of $\beta$ -Chloroethylamines Containing Heterocyclic Nuclei

BY KENNETH N. CAMPBELL, JOSEPH F. ACKERMAN<sup>1</sup> AND BARBARA K. CAMPBELL

In view of the effectiveness of dibenzylaminoethyl chloride hydrochloride "Dibenamine"<sup>2</sup> as a sympatholytic drug,<sup>3</sup> it became of interest to prepare analogous substances in which one of the phenyl groups is replaced with a heterocyclic ring system. In this paper we are reporting the synthesis of five such compounds



All of these compounds were prepared from the corresponding amino alcohols by the action of thionyl chloride, with or without an inert solvent. The intermediate amino alcohols were made in one of the two following ways



The thiophene compound (I) was prepared by method A from  $\alpha$ -chloromethylthiophene without difficulty. In the case of the furan derivative (II) it was found more convenient to use the aldehyde with benzylamine than to prepare  $\alpha$ -chloromethylfuran; this synthesis likewise presented no difficulty.

When attempts were made to use 4-chloromethylimidazole in Method A for the synthesis of compound III, it was found very difficult to purify the imidazolylmethylbenzylamine from unreacted benzylamine, as the mixture could not be distilled, and the two substances had about the same solubilities. The amino alcohol was, therefore, prepared by Method B, and the crude amino alcohol was converted to the chloride (III) without purification. Compound III was separated from the benzylaminoethyl chloride hydrochloride which accompanied it by recrystallization from methanol.

It was originally planned to prepare the quino-

line compound (IV) from cinchoninic aldehyde and benzylamine, by Method A. Cinchoninic aldehyde has been condensed successfully with other primary amines, and the aldimines reduced to secondary amines<sup>4,5</sup> but in the present case only poor yields of lepidylbenzylamine could be obtained by this procedure. The action of N-bromosuccinimide on lepidine was, therefore, investigated. Buu-Hoi<sup>6</sup> has reported that quin-aldine and  $\alpha$ - and  $\gamma$ -picolines are very easily brominated by this reagent, but he did not describe the products in detail nor establish their structures. We have found that lepidine reacts readily with N-bromosuccinimide in carbon tetrachloride, but the product is very unstable. It decomposes rapidly in hot solvents, and fairly rapidly even on standing, so that we were unable to purify it or analyze it. The product must be

$\alpha$ -bromomethylquinoline, however, since it reacts rapidly with benzylaminoethanol, and behaves in other respects also as a reactive bromide. The N-lepidyl-N-benzylaminoethanol was converted, without difficulty, to the chloride (IV).

The thiazolyl compound (V) was prepared by Method B. Both the amino alcohol and the chloride in this series formed very hygroscopic salts, which made purification difficult; the oxalate of the chloride was found to be more easily handled than the hydrochloride.

Attempts were made to prepare a pyrimidine derivative by Method B, from 2-methyl-4-amino-5-bromomethylpyrimidine, but it was difficult to separate the reaction products from starting material and no satisfactory drug was obtained.

The authors wish to thank the Smith, Kline and French Co. of Philadelphia for a fellowship grant to support this work, and Dr. Fellows of Smith, Kline and French Co. for making the pharmacological tests. Most of the compounds

(1) Smith, Kline and French Fellow, 1947-1948.  
 (2) "Dibenamine" is the trade-mark for dibenzyl-3-chloroethylamine hydrochloride, Smith, Kline and French Laboratories.

(3) Nickerson, Goodman and Nomaguchi, *J. Pharmacol.*, **89**, 167 (1947).

(4) Campbell, Sommers, Kerwin and Campbell, *THIS JOURNAL*, **68**, 1851 (1946).

(5) Phillips, *ibid.*, **69**, 865 (1947).

(6) Buu-Hoi, *Ann.*, **556**, 5 (1944).