bromoanisole. To this solution was added 21.0 g. (0.11 mole) of 9-anthrone in 100 ml. of tetrahydrofuran. After stirring for 45 min., the mixture was poured into ice–concentrated hydrochloric acid. The organic products were extracted with benzene and the extracts were dried over sodium sulfate. Evaporation of the benzene left an oily residue that was chromatographed on alumina in 3:2 benzene–hexane. The blue-fluorescing band was collected and recrystallized from aqueous ethanol to yield 7.4 g. (24%) of pale yellow product: m.p. 113.5–114.5°; $\lambda_{\max}^{\text{ethanol}}$ mμ (log ε), 254 (5.16), 346 (3.99), 364 (4.03), and 384 (3.83).

Anal. Calcd. for $C_{21}H_{16}O$: C, 88.70; H, 5.67. Found: C, 88.63; H, 5.67.

General Procedure for Preparation of 9-Methoxyphenyl-10-nitrophenylanthracenes.—A solution of 1.42 g. (5.0 mmoles) of a 9-methoxyphenylanthracene[7] and 0.83 g. (5.0 moles) of cupric chloride dihydrate in 240 ml. of acetone was added to a three-necked flask fitted with dropping funnel, condenser, and mechanical stirrer. The system was flushed with nitrogen and to the flask was added 60 ml. of an aqueous solution of a nitrobenzenediazonium chloride, prepared from 3.45 g. (25 mmoles) of nitroaniline, 12 ml. of 6 *N* hydrochloric acid, 1.73 g. (25 mmoles) of sodium nitrite, and diluted with water to volume. After stirring for about 1 hr. (9-*o*-nitrophenyl- and 9-*p*-nitrophenyl-10-*p*-methoxyphenylanthracene crystallize from reaction mixtures and may be isolated by filtration), the reaction mixture was poured into 500 ml. of water and the organic products were extracted with three 50-ml. portions of benzene. The combined extracts were washed with 20% aqueous sodium hydroxide and with water. After drying over sodium sulfate, the benzene was removed under reduced pressure and codistillation with ethylene glycol was employed to remove nitrochlorobenzene. The distillate was discarded and the ethylene glycol solution of the nonvolatile fraction was diluted with water and gave a viscous oil that was dissolved in benzene. After drying over anhydrous sodium sulfate, the benzene was removed under reduced pressure. Chromatography of the residue in 2:1 benzene–hexane on 100 g. of neutral alumina (Merck) gave 9-methoxyphenylanthracene followed by a yellow band of 9-methoxyphenyl-10-nitrophenylanthracene. The yellow products were recrystallized from benzene–ethanol or benzene–hexane. Yields, melting points, and elemental analyses of the nine constitutional isomers are listed in Table I and the ultraviolet spectra are recorded in Table II.

The three reverse syntheses from diazotized anisidines and 9-nitrophenylanthracenes[1] were accomplished as described above except that 10 equiv. of diazonium salt was used and the reactions were continued for 2 hr. The yields are recorded in Table I.

General Procedure for Preparation of 9-Phenyl- and 9-Nitrophenyl-10-nitrophenylanthracenes.—A solution of 0.254 g. (1 mmole) of 9-phenylanthracene or 0.299 g. (1 mmole) of 9-nitrophenylanthracene[1] and 0.170 g. (1 mmole) of cupric chloride dihydrate in 40 ml. of acetone was placed in a three-necked flask fitted with a stirrer, condenser, and addition funnel. The apparatus and contents were flushed with nitrogen and to the flask was added with stirring 10 ml. of an aqueous solution of nitrobenzenediazonium chloride that had been prepared from 1.04 g. (7.5 mmoles) of nitroaniline, 1.87 ml. of concentrated hydrochloric acid, diluted with 1.5 ml. of water, and 0.569 g. (8.2 mmoles) of sodium nitrite. The excess nitrous acid was destroyed with sulfamic acid and the solution was diluted to volume with water. After addition of the solution of diazonium chloride, the stirring was continued for 30 min. and the reaction mixture was refrigerated overnight. 9-Phenyl-10-*p*-nitrophenylanthracene, 9-phenyl-10-*o*-nitrophenylanthracene, 9-*o*-nitrophenyl-10-*p*-nitrophenylanthracene, and 9-*m*-nitrophenyl-10-*p*-nitrophenylanthracene usually crystallized from the respective reaction mixtures and were isolated by filtration. In the event that precipitation did not occur, the reaction mixture was steam distilled and the distillate was discarded. The nonvolatile residue was dissolved in benzene and the benzene solution was concentrated and dried by distillation. Chromatography of this solution on alumina (Merck), prepared and eluted with 1:1 benzene–hexane, gave 9-phenylanthracene or 9-nitrophenylanthracene followed by a yellow band of 9-phenyl-10-nitrophenylanthracene or 9-nitrophenyl-10-nitrophenylanthracene. Recrystallization from benzene–hexane and/or benzene–ethanol afforded analytical samples. However, 9-*o*-nitrophenyl-10-*m*-nitrophenylanthracene yields a molecular compound of m.p. 218° dec. when crystallized from benzene and the higher melting point, 244–246°, refers to material recrystallized from ethanol. The reverse syntheses were accomplished by an essentially identical procedure. Yields, melting points, and elemental analyses are recorded in Table I and ultraviolet data are recorded in Table II.

9-*p*-Chlorophenyl-10-*p*-nitrophenylanthracene.—This compound was prepared from 9-*p*-nitrophenylanthracene and diazotized *p*-chloroaniline and from 9-*p*-chlorophenylanthracene and diazotized *p*-nitroaniline in the manner described previously for 9-nitrophenyl-10-nitrophenylanthracenes. Both syntheses furnished solid products directly from the respective reaction mixtures. Recrystallization from benzene–ethanol afforded an analytical sample. Pertinent data are listed in Tables I and II.

9-*p*-Chlorophenyl-10-*p*-nitrophenylanthracene may also be prepared directly from anthracene without isolation of the 9-arylanthracene. For example, anthracene was treated with *p*-nitrobenzenediazonium chloride by the procedure previously described[1] and the 9,10-di-*p*-nitrophenylanthracene that precipitated from the reaction mixture was removed by filtration. The filtrate was then treated with diazotized *p*-chloroaniline and the resulting yellow precipitate was isolated by filtration. The yield based on anthracene with 5 equiv. of each diazonium salt amounted to 6%.[8]

Infrared Spectra.—The infrared spectra of these compounds are complex. For example, 9-*p*-methoxyphenyl-10-*o*-nitrophenylanthracene exhibits more than twenty strong absorption bands between 6 and 15 μ. However, all nineteen of these derivatives of 9,10-diphenylanthracene (1–19) exhibit absorption bands, characteristic of the nitro group, at 6.5–6.7 and at 7.4–7.5 μ. In compounds 10–18, aryl methyl ether absorption bands appear at 8.0–8.2 and at 9.5–9.7 μ.

Acknowledgment.—The authors are indebted to the Aeronautical Research Laboratory, Wright-Patterson Air Force Base, for financial support and to Mr. Derek De Souza for measuring the ultraviolet spectra.

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[6] O. Diels and F. Bunzl, *Ber.*, **38**, 1486 (1905).

[7] The synthesis of 9-*p*-methoxyphenylanthracene has been reported in ref. 1.

[8] This experiment was performed by Dr. Leon B. Levy.

The Reaction of Substituted Ureas with Lithium Aluminum Hydride. I. A New Synthesis of Three-Substituted Formamidines

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Received March 24, 1964

As far as we know, the only previous attempt to reduce ureas with lithium aluminum hydride (LAH) was carried out by Ried and Muller,[1] who, after heating an ether solution of LAH with *sym*-diphenylurea for 30 hr., did not obtain any reaction products.

We have now found that LAH does react with other ureas. In fact, after heating 1,1-diethyl-3-phenylurea with equimolar quantities of LAH in benzene–ether solutions for 14 hr., two fractions could be isolated, besides unreacted urea. The first one was easily recognized as N-methylaniline, the other one was identified as N-phenyl-N′,N′-diethylformamidine (I), on the basis of microanalysis and of ultraviolet and infrared spectra. The ultraviolet spectrum,[2] with one maximum at 265 mμ (log ε 4.12), suggested the formation of a double bond conjugated to the benzene ring, as a consequence of the action of LAH on the urea. The in-

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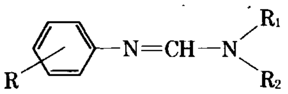
[1] W. Ried and F. Muller, *Chem. Ber.*, **85**, 470 (1952).

[2] This was determined with a Beckman (Model DK) recording instrument in 95% ethanol.

TABLE I

DISUBSTITUTED N-ARYLFORMAMIDINES

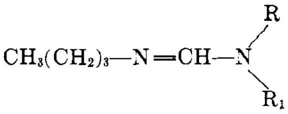


No.	R	R_1	R_2	B.p., °C. (mm.)	Procedure	Yield, %	Calcd., %			Found, %			Picrate, m.p., °C.
							C	H	N	C	H	N	
1	H	CH_3	CH_3	128–131[a] (15)	B (C, A)	29 (28, 26)	72.94	8.16	18.90	73.11	8.29	19.05	158
2	H	C_2H_5	C_2H_5	143–144[b] (15)	A	57	74.95	9.15	15.90	75.10	9.02	15.64	140–141
3	H	$CH_3(CH_2)_2$	$CH_3(CH_2)_2$	154–155 (15)	A	62	76.42	9.87	13.71	76.68	10.00	13.59	130–131
4	H	$(CH_3)_2CH$	$(CH_3)_2CH$	c	A	57	76.42	9.87	13.71	76.13	9.78	13.82	156–157
5	H	$CH_3(CH_2)_3$	$CH_3(CH_2)_3$	102–104[d] (0.001)	A	53	77.53	10.41	12.06	77.71	10.30	12.11	127
6	H	$(CH_3)_2CHCH_2$	$(CH_3)_2CHCH_2$	93–95 (0.001)	A	40	77.53	10.41	12.06	77.80	10.41	12.22	129–130
7	H	CH_3	C_6H_{11}[e]	124–125 (0.001)	A	40	77.73	9.32	12.95	77.50	9.27	13.02	170
8	H	$CH_2CH_2CH_2CH_2$		115–117[f] (0.05)	B (A)	35 (traces)	75.82	8.10	16.08	75.60	8.14	16.11	191–192
9	H	$CH_2CH_2CH_2CH_2CH_2$		115–118 (0.06)	B (A)	36 (traces)	76.55	8.57	14.88	76.67	8.32	14.60	149–150
10	H	$CH_2CH_2OCH_2CH_2$		120[g] (0.05)	B (A)	40 (6)	69.44	7.42	14.73	69.56	7.31	14.90	181–182
11	H	$CH_2CH_2N(CH_3)CH_2CH_2$		100–102[h] (0.001)	C (A)	43 (12)	70.90	8.43	20.67	70.61	8.15	20.44	215[i]
12	2-Cl	C_2H_5	C_2H_5		A								112–113[j]
13	3-Cl	C_2H_5	C_2H_5	108 (0.001)	A	52	62.64	7.17	13.30	62.82	7.01	13.08	142
14	4-Cl	C_2H_5	C_2H_5	166–167 (15)	A	61	62.64	7.17	13.30	62.22	6.84	13.12	180
15	4-OC_2H_5	C_2H_5	C_2H_5	114–115 (0.005)	A	60	70.87	9.15	12.72	70.58	9.18	12.60	153–154
16	2-NH_2	C_2H_5	C_2H_5		A								186[i,k]

[a] M. L. Weiner [*J. Org. Chem.*, **25**, 2245 (1960)] gives b.p. 68–71° at 0.05 mm. [b] W. J. Comstock and H. L. Wheeler [*Am. Chem. J.*, **13**, 519 (1891); F. K. Beilstein, "Handbuch der organischen Chemie," Vol. 12, p. 236] gives b.p. 272–275°. [c] M.p. 56–58°, from petroleum ether (b.p. 60–68°). [d] Lit.[3] b.p. 190–192° at 16 mm. [e] Cyclohexyl. [f] E. Enders [German Patent 952,631 (Nov. 22, 1956); *Chem. Abstr.*, **53**, 7197*i* (1959)] reports b.p. 170–176° (16 mm.). [g] M.p. 65–66°, from petroleum ether (b.p. 60–68°). [h] M.p. 39–40°, from petroleum ether (b.p. 60–68°). [i] Dipicrate. [j] *Anal.* Calcd. for $C_{17}H_{18}ClN_5O_7$: C, 46.42; H, 4.12; N, 15.90. Found: C, 46.63; H, 4.28; N, 15.96. [k] *Anal.* Calcd. for $C_{23}H_{23}N_9O_{14}$: C, 42.49; H, 3.57; N, 19.39. Found: C, 42.44; H, 3.18; N, 19.62.

TABLE II

DISUBSTITUTED N-BUTYLFORMAMIDINES



$CH_3(CH_2)_3—N=CH—N(R)(R_1)$

No.	R	R₁	B.p., °C. (mm.)	Procedure	Yield, %	Calcd., % C	Calcd., % H	Calcd., % N	Found, % C	Found, % H	Found, % N	Picrate,[a] m.p., °C.
1	C_2H_5	C_2H_5	75–76 (15)	D (A)	51 (17)	69.17	12.90	17.93	69.32	12.86	17.80	54–55
2	$CH_3(CH_2)_2$	$CH_3(CH_2)_2$	92–94 (15)	D (A)	48 (14)	71.68	13.13	15.20	71.53	13.10	15.25	63–64
3	$(CH_3)_2CH$	$(CH_3)_2CH$	87–88(15)	D (A)	50 (13)	71.68	13.13	15.20	71.59	13.22	15.16	84
4	$CH_3(CH_2)_3$	$CH_3(CH_2)_3$	124–126 (17)	D (A)	53 (13)	73.52	13.29	13.20	73.28	13.40	12.93	49–50
5	CH_3	C_6H_{11}[b]	129–130 (15)	A	28	73.41	12.32	14.27	73.27	12.50	14.19	75–76
6	$CH_2CH_2CH_2CH_2$		97–98 (15)	A (D)	38 (33)	70.07	11.76	18.16	69.80	11.50	18.40	c
7	$CH_2CH_2CH_2CH_2CH_2$		103–104 (15)	D (A)	51 (35)	71.37	11.98	16.65	71.05	12.25	16.42	53–54
8	$CH_2CH_2OCH_2CH_2$		107–110 (15)	E (A)	28 (14)	63.49	10.66	16.46	63.66	11.00	16.25	84–85
9	$CH_2CH_2N(CH_3)CH_2CH_2$		110–111 (15)	D (A)	39 (38)	65.52	11.55	22.93	65.18	11.47	23.21	171–172[d]

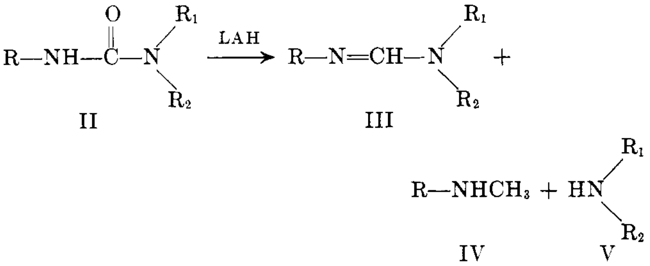
[a] The picrates were obtained from an ether solution. [b] Cyclohexyl. [c] We had not succeeded in crystallizing this picrate. [d] Dipicrate.

frared spectrum supported this hypothesis, showing an intense absorption band at 6.10 and no absorption band in the 2.90–3.90-μ region.

The structure of the reaction product was conclusively confirmed by comparison with the formamidine (I) prepared according to a standard method.[3]

In order to extend this new reaction, we treated with LAH other ureas, corresponding to the general formula II, according to Scheme I.

SCHEME I



R = butyl or aryl group

R_1 and R_2 = alkyl, $(-CH_2-)_n$, or $-(CH_2CH_2 \ldots CH_2CH_2)-$

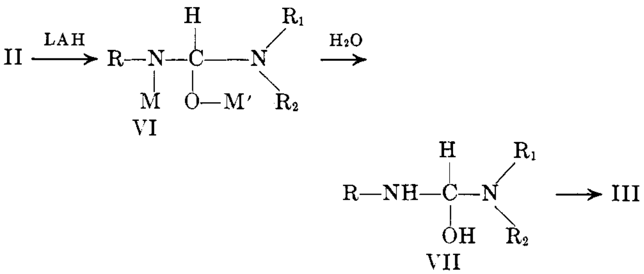
In order to make the results of the new tests as comparable with one another as possible, the reactions were carried out under standard conditions except as noted below. These conditions, which were chosen on the basis of preliminary tests with 1,1-diethyl-3-phenylurea, involved refluxing a 1:1 benzene–ether solution of the urea–lithium aluminum hydride mixture (molar ratio of 2:3) for 14 hr.

The reaction of Scheme I usually gives, in addition to the expected formamidine III, a mixture of the amines derived from the cleavage of the C–N$_t$ bond, *viz.*, N-methylaryl- or butylamine (IV) and the secondary amine V. If the reaction conditions were too drastic, a high yield of amines was observed; when the conditions were too mild, most of the initial urea was recovered unchanged.

The standard conditions appeared to be too drastic for the phenylureas when the tertiary nitrogen was part of a ring (high amine yield), whereas they appeared to be too mild for the 1,1-dialkyl-3-butylureas (high percentage of unreacted urea). In either case appropriate changes in the reaction conditions gave more satisfactory formamidine yields (see Tables I and II). In neither case, however, could the high formamidine yields obtained from the 1,1-dialkyl-3-arylureas in the standard conditions (50–60%, except for 1,1-dimethyl-3-phenylurea) be reached.[4]

On the basis of the mechanisms[5,6] already suggested for LAH reductions of amides, the formation of trisubstituted formamidines from the corresponding ureas can be explained (see Scheme 2) by assuming that the reaction between ureas and LAH stops, partly, at the intermediate complex VI, *i.e.*, that the C–N bonds neighboring the C–O bond enhance the resistance to cleavage of the latter. Upon hydrolysis, VI would give formamidine, by way of the unstable α-amino alcohol VII.

SCHEME II



M = AlLi/4

M′ = AlH_3^- or $Al^-/4$

The formation of secondary amines, which almost always accompanies the formation of formamidines, can be explained by considering that the above mechanism competes with that involving the cleavage of the C–O bond and/or of the C–N bond in VI. We note that in the first case the amines should originate from a methanediamine derivative, which would be further decomposed by LAH with cleavage of its C–N$_t$ bond.[7,8]

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[3] H. G. Mandel and A. J. Hill, *J. Am. Chem. Soc.*, **76**, 3978 (1954).

[4] It was not possible to obtain the corresponding formamidines when a phenyl group was present on the trisubstituted nitrogen atom; the only products isolated were the appropriate amines and, depending upon the reaction conditions, the starting urea.

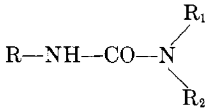
[5] V. M. Mićović and M. L. Mihailović, *Serbian Acad. Sci. Monographs*, **237**, 58 (1955).

[6] N. G. Gaylord, "Reduction with Complex Metal Hydrides," Interscience Publishers, Inc., New York, N. Y., 1956, pp. 544–546.

[7] This suggestion is supported by some experiments carried out on 4-(anilinomethyl)morpholine[8] (VIII). In fact, by heating ether solutions of 0.02 mole of VIII with 0.02 and 0.015 mole of LAH, respectively, for 14 hr., we were able to isolate only N-methylaniline and morpholine from the reaction mixtures.

[8] M. Zief and J. F. Mason, *J. Org. Chem.*, **8**, 1 (1943).

TABLE III
DISUBSTITUTED BUTYL- OR ARYLUREAS



$R{-}NH{-}CO{-}N(R_1)(R_2)$

R	R_1	R_2	Yield, %	M.p., °C.	B.p., °C. (mm.)	Calcd., % C	Calcd., % H	Calcd., % N	Found, % C	Found, % H	Found, % N
$CH_3(CH_2)_3$	C_2H_5	C_2H_5	87	. . .	104–105 (0.001)	62.74	11.70	16.26	62.81	11.71	16.20
$CH_3(CH_2)_3$	$CH_3(CH_2)_2$	$CH_3(CH_2)_2$	78	. . .	109–110 (0.001)	65.95	12.08	13.99	65.99	12.12	13.91
$CH_3(CH_2)_3$	$(CH_3)_2CH$	$(CH_3)_2CH$	90	. . .	128–130 (15)	65.95	12.08	13.99	65.70	12.15	13.90
$CH_3(CH_2)_3$	CH_3	C_6H_{11}[a]	80	. . .	135–138 (0.005)	67.88	11.39	13.19	68.16	11.60	13.32
$CH_3(CH_2)_3$	$CH_2CH_2CH_2CH_2$		91	52–53[b]	. . .	63.49	10.66	16.46	63.67	10.78	16.59
$CH_3(CH_2)_3$	$CH_2CH_2CH_2CH_2CH_2$		87	63–64[b]	. . .	65.17	10.94	15.20	65.31	11.02	14.97
$CH_3(CH_2)_3$	$CH_2CH_2OCH_2CH_2$		90	68[b]	. . .	58.03	9.74	15.04	57.74	9.62	15.20
$CH_3(CH_2)_3$	$CH_2CH_2N(CH_3)CH_2CH_2$		86	54–56[b]	112–115 (0.001)	60.26	10.62	21.08	60.32	10.48	21.04
C_6H_5	$CH_3(CH_2)_2$	$CH_3(CH_2)_2$	85	73[c]	. . .	70.87	9.15	12.72	70.82	9.12	12.86
4-$C_2H_5OC_6H_4$	C_2H_5	C_2H_5	86	95–96[c]	. . .	66.07	8.53	11.86	65.80	8.54	11.72

[a] Cyclohexyl. [b] Crystallized from petroleum ether, (60–68°). [c] Crystallized from a benzene-petroleum ether mixture.

Experimental[9]

Preparation of Ureas.—1,1-Diethyl-3-(*o*-aminophenyl)urea was prepared by catalytic reduction of the corresponding nitro compound. The other ureas used were prepared according to a previously reported method.[10] The previously unknown ureas are listed in Table III.

Preparation of Formamidines. Procedure A.—A solution of 0.02 mole of the urea in 150 ml. of anhydrous benzene was added, dropwise, to a well-stirred suspension of 0.03 mole of LAH in 150 ml. of dry ether. The reaction mixture was refluxed for 14 hr. Then, the mixture was cooled and the reaction product and the excess LAH were decomposed by slow addition of water, 15% aqueous NaOH, and water, in succession.[11] After vigorous stirring for 20 min., the mixture was filtered and the precipitate was repeatedly washed with ether. The initial solution and the ether used for washing were combined and shaken with a concentrated solution of tartaric acid. The organic phase, after evaporation of the solvent, gave the unreacted urea; the aqueous phase, after cooling, was made alkaline with potassium carbonate and extracted with ether. The resulting ether solution was dried on anhydrous sodium sulfate, and then evaporated. The residue was then distilled under reduced pressure.

Procedure B.—A solution of 0.02 mole of the urea in 150 ml. of dry benzene was added, dropwise and with stirring, to a suspension of 0.02 mole of LAH in 150 ml. of anhydrous ether. When the addition was finished, the mixture was heated under reflux for 14 hr. The cooled mixture was, successively, worked up as in procedure A.

Procedure C.—This was identical with procedure A, except for the refluxing time (4 hr.).

Procedure D.—A solution of 0.05 mole of the urea in 100 ml. of toluene was added, slowly and under stirring, to a suspension of 0.05 mole of LAH in 100 ml. of *n*-butyl ether. After the addition was complete, the mixture was refluxed for 5 hr. After cooling, the reaction mixture was decomposed and worked up as in procedure A.

Procedure E.—This was similar to procedure B and differed only in the heating time (4 hr.).

Acknowledgment.—The authors would like to thank Dr. G. Del Re, Professor of Theoretical Chemistry at the University of Naples, for several useful discussions. The help of Drs. A. De Leonibus and M. L. Reviglio in obtaining the microanalysis data and spectra is also acknowledged.

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[9] Melting points were determined on a Fisher-Johns apparatus and are uncorrected.

[10] W. Gebhardt, *Chem. Ber.*, **17**, 3039 (1884).

[11] V. M. Mićović and Mihailović, *J. Org. Chem.*, **18**, 1196 (1953).

Proton Magnetic Resonance Evidence for Ligand–Porphyrin Interaction in Magnesium Porphyrins[1]

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Recently Corwin, *et al.*,[3,4] proposed that the appearance of a new absorption peak in the near ultraviolet (Soret) region of the electronic spectra of metalloporphyrins in the presence of strong ligands is due to a steric interference between the ligand and the π-electron system of the porphyrin ring. Caughey and Iber[5] recently showed that in N-methyl and N-ethyletioporphyrin II (etio II) the position of the N-alkyl resonance is shifted to considerably higher field due to the ring-current effects at the center of the aromatic porphyrin system. We have observed large chemical shifts to high field from their normal position in the case of ligands bound to magnesium in magnesium etioporphyrin II (Mg etio II) and magnesium tetraphenylporphin (Mg TPP). This permits one to determine the number of ligands interacting with the metalloporphyrin and to gain a qualitative view of how tightly the ligand is bound to the metalloporphyrin system.

Table I presents a comparison of the chemical shifts of pyridine in the Mg etio II complex, Mg TPP complex, and some model pyridine complexes. In both the pyridinium hydrochloride and the magnesium hexapyridinate diiodide the pyridine protons are shifted to slightly lower field, a paramagnetic shift expected from the de-

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[1] Porphyrin studies XXXIII. Paper XXXII: E. W. Baker, M. S. Brookhart, and A. H. Corwin, *J. Am. Chem. Soc.*, **86**, 4587 (1964).

[2] National Institutes of Health Predoctoral Fellow, 1962–1964.

[3] A. H. Corwin, D. G. Whitten, E. W. Baker, and G. G. Kleinspehn, *J. Am. Chem. Soc.*, **85**, 3621 (1963).

[4] D. G. Whitten, E. W. Baker, and A. H. Corwin, *J. Org. Chem.*, **28**, 2363 (1963).

[5] W. S. Caughey and P. K. Iber, *ibid.*, **28**, 269 (1963).

[6] F. W. Sears and M. W. Zemansky, "University Physics," Addison-Wesley Publishing Co., Inc., Reading, Mass., 1949, p. 594.