1,4-Cycloaddition Reactions. III. Synthesis of Furo[3,2-*c*]pyrido[2,3-*g*]quinolines, Furo[2,3-*a*][4,7]phenanthrolines, Furo[3,2-*c*]pyrrolo[2,3-*g*]quinolines, Furo[3,2-*c*]pyrrolo[3,2-g]quinolines, and Furo[3,2-*c*]furo[2′,3′:4,5]pyrido[2,3-*g*]quinolines from 2,3-Dihydro-5-methylfuran and Schiff Bases (1)

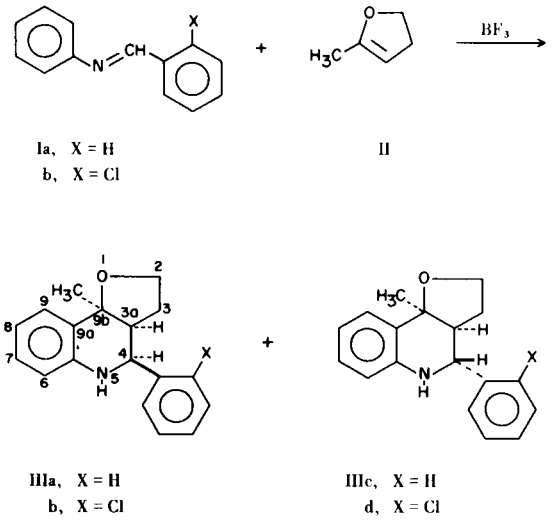
S. C. Perricone, Donald F. Worth, and Edward F. Elslager

Chemistry Department, Division of Medical and Scientific Affairs, Parke, Davis and Company

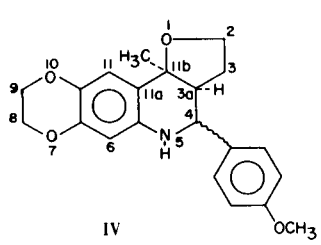
The 1,4-cycloaddition of 2,3-dihydro-5-methylfuran (II) to 1-acetyl-1,2,3,4-tetrahydro-6-[(*p*-hydroxybenzylidene)amino]quinoline (VIII) in the presence of boron trifluoride gave two pairs of epimers, namely *dl*-10-acetyl-2,3,3a,4,5,7,8,9,10,11b-decahydro-4-(*p*-hydroxyphenyl)-11b-methylfuro[3,2-*c*]pyrido[2,3-*g*]quinoline (IXa and b) and *dl*-8-acetyl-2,3,3a,4,5,8,9,10,11,-11c-decahydro-4-(*p*-hydroxyphenyl)-11c-methylfuro[2,3-*a*][4,7]phenanthroline (Xa and b). *dl*-9-Acetyl-3,3a,4,5,7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-c]pyrrolo-[2,3-*g*]quinoline (XIIIa) was the predominant product isolated from the reaction of II with 1-acetyl-5-[*p*-(hydroxybenzylidene)amino]indoline (XII). When 1-acetyl-6-[(*p*-hydroxybenzylidene)amino]indoline (XVI) was treated with 2,3-dihydro-5-methylfuran (II), two epimers of *dl*-7-acetyl-3,3a,4,5,7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-*c*]pyrrolo[3,2-*g*]quinoline (XVIIa and b) were obtained. *dl*-2,3,3a,4,5,6b,8,9,9a,10,11,12b-Dodecahydro-4,10-bis(*p*-methoxyphenyl)-6b,12b-dimethylfuro[3,2-*c*]furo[2′,3′:4,5]pyrido[2,3-*g*]quinoline (XX) was formed when 2,3-dihydro-5-methylfuran was allowed to react with *N*,*N*′-bis(*p*-methoxybenzylidene)-*p*-phenylenediamine (XIX). Structure assignments were made from NMR spectra. None of the compounds exhibited appreciable biological activity.

Povarov and co-workers recently developed an elegant quinoline synthesis utilizing the cycloaddition of vinyl ethers to Schiff bases (2). Moreover, the condensation of the cyclic enol ether 2,3-dihydro-5-methylfuran (II) with *N*-benzylideneaniline (Ia) led to the formation of the interesting tricyclic derivative 2,3,3a,4,5,9b-hexahydro-9b-methyl-4-phenylfuro[3,2-*c*]quinoline in high yield (3). A reinvestigation of the latter reaction in these laboratories showed that the reaction of 2,3-dihydro-5-methylfuran (II) with either *N*-benzylideneaniline (Ia) or *N*-(*o*-chlorobenzylidene)aniline (Ib) (Chart I) did not yield a single product but instead gave two epimers (IIIa and c and IIIb and d, respectively) in approximately equal amounts (4).

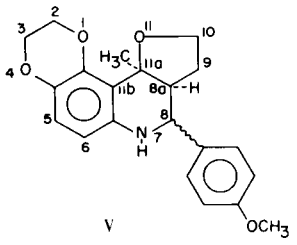
CHART I



In view of the antimalarial activity exhibited by *dl*-2,3,3a,4,5,9b-hexahydro-9b-methyl-4-phenylfuro[3,2-*c*]-quinoline against ***Plasmodium berghei*** in the mouse (4), further work was initiated in these laboratories directed toward the synthesis and structure determination of other novel heterocyclic compounds derived from 2,3-dihydro-5-methylfuran and Schiff bases (1). Thus, the boron trifluoride-catalyzed 1,4-cycloaddition of 2,3-dihydro-5-methylfuran to *N*-(*p*-methoxybenzylidene)-1,4-benzodioxan-6-amine gave 2 pairs of epimers, *dl*-2,3,3a,4,5,8,9,11b-octahydro-4-(*p*-methoxyphenyl)-11b-methyl-*p*-dioxino-[2,3-*g*]furo[3,2-c]quinoline (IV, 3a,4-protons *cis* and *trans*) and *dl*-2,3,7,8,8a,9,10,11a-octahydro-8-(*p*-methoxyphenyl)-11a-methyl-*p*-dioxino[2,3-*f*]furo[3,2-*c*]quinoline

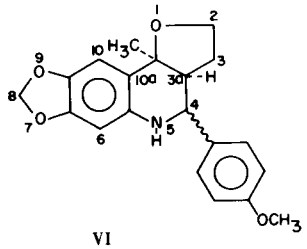


IV



V

(V, 8,8a-protons *cis* and *trans*) (1). For isomers of like configuration, the NMR signals from the angular methyl groups were further downfield for compounds containing the angular ring system (V) compared to the linear ring system (IV). Further, the peaks from the isomers with the 3a,4- or 8a,8-protons in the *cis* configuration were downfield with respect to those from the corresponding *trans* isomers. A similar pattern was seen in a series of *dl*-2,3,3a,4,5,12d(and 12b)hexahydro-4-phenyl-12d(and 12b)methylbenzofuro[3,2-*f*(and 2,3-*g*)]furo[3,2-*c*]quinolines (5). When *N*-(*p*-methoxybenzylidene)-3,4-methylenedioxyaniline was condensed with II in an analogous manner, a mixture of 2 epimers of *dl*-2,3,3a,4,5,10b-hexahydro-4-(*p*-methoxyphenyl)-10b-methyl[1,3]dioxolo-[4,5-*g*]furo[3,2-*c*]quinoline (VI, 3a,4-protons *cis* and *trans*) was isolated (1). Unfortunately, compounds IV-VI lacked appreciable antimalarial activity against *P. berghei* in the mouse (1).



VI

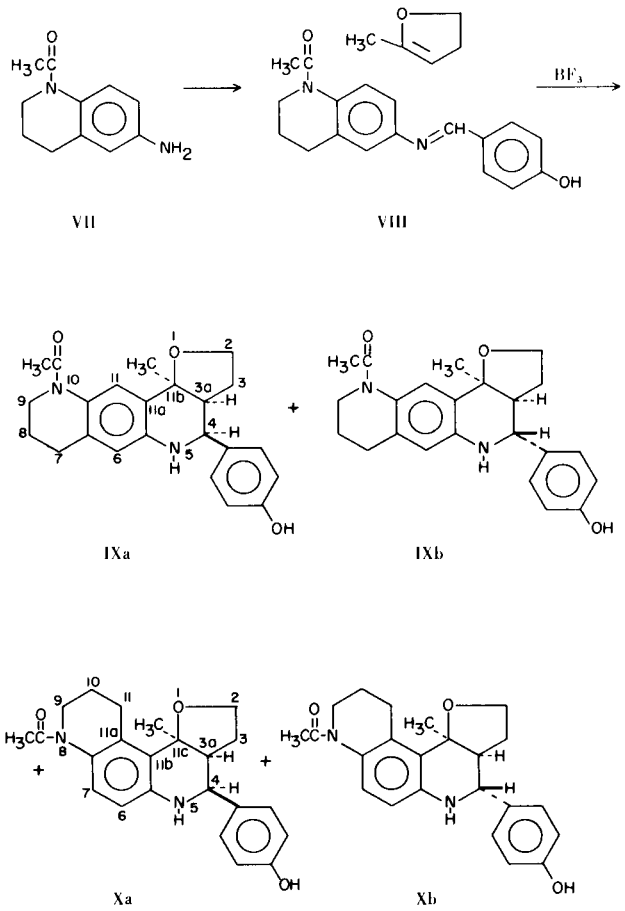
The present communication describes five additional heterocyclic systems that have been prepared by the cycloaddition of 2,3-dihydro-5-methylfuran to Schiff bases derived from 1-acetyl-6-amino-1,2,3,4-tetrahydroquinoline (VII), 1-acetyl-5-aminoindoline (XI), 1-acetyl-6-aminoindoline (XV), and *p*-phenylenediamine, namely furo-[3,2-*c*]pyrido[2,3-*g*]quinolines (IXa and b, Chart II), furo[2,3-*a*][4,7]phenanthrolines (Xa and b, Chart II), furo[3,2-*c*]pyrrolo[2,3-*g*]quinolines (XIIIa and b, Chart III), furo[3,2-*c*]pyrrolo[3,2-*g*]quinolines (XVIIa and b, Chart IV), and furo[3,2-*c*]furo[2',3':4,5]pyrido[2,3-*g*]-quinolines (XX, Chart V). These ring systems are not listed in *Chemical Abstracts* or "The Ring Index" (6) and appear to be novel heterocyclic types.

As in previous work (1-5), elemental analyses of the purified isomer mixtures isolated from each reaction showed that 1:1 adducts had been obtained. However, separation of these isomers was not undertaken in the present studies because the mixtures lacked appreciable biological activity. Tentative structure assignments have been made on the basis of the NMR spectra (7).

The cycloaddition of 2,3-dihydro-5-methylfuran (II) to the *N*-heterocyclic Schiff bases VIII, XII, and XVI proceeded more sluggishly than with previously reported systems (1-5). Excess 2,3-dihydro-5-methylfuran and multiple charges of boron trifluoride etherate catalyst were usually required. The initial introduction of a large excess of boron trifluoride was avoided in the fear that substantial amounts of polymeric 2,3-dihydro-5-methylfuran would be formed leading to difficulties in the isolation of the desired products.

It was anticipated that the cycloaddition of 2,3-dihydro-5-methylfuran (II) to 1-acetyl-1,2,3,4-tetrahydro-6-[(*p*-hydroxybenzylidene)amino]quinoline (VIII) might give four products, namely *dl*-10-acetyl-2,3,3a,4,5,7,8,9,-10,11b-decahydro-4-(*p*-hydroxyphenyl)-11b-methylfuro-[3,2-*c*]pyrido[2,3-*g*]quinoline (IXa and b) and *dl*-8-acetyl-2,3,3a,4,5,8,9,10,11,11c-decahydro-4-(*p*-hydroxyphenyl)-

CHART II



VII VIII

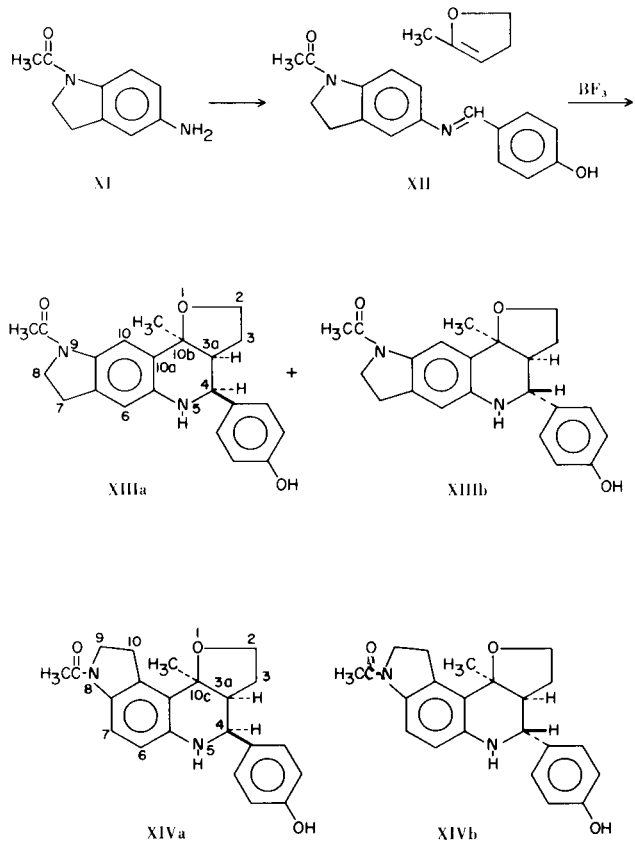
IXa IXb

Xa Xb

11c-methylfuro[2,3-*a*][4,7]phenanthroline (Xa and b) (Chart II). The reaction proceeded sluggishly and required several equivalents of the vinyl ether II and periodic additions of the boron trifluoride catalyst for completion. The product that was isolated and purified (48% yield) showed 3 spots on TLC (silica; ethyl acetate, triethylamine). However, the NMR spectrum of this material exhibited peaks at 1.98, 1.58, 1.31, and 1.25. It is therefore proposed that this material, in concordance with IV and V, is a mixture of all four isomers with 11b-CH_3 absorption at 1.58 (IXa) and 1.25 (IXb) and 11c-CH_3 absorption at 1.98 (Xa) and 1.31 (Xb) (Chart II).

By analogy, the reaction of 2,3-dihydro-5-methylfuran (II) with 1-acetyl-5-[(*p*-hydroxybenzylidene)amino]indoline (XII) might be expected to give *dl*-9-acetyl-3,3a,4,5,-7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-*c*]pyrrolo[2,3-*g*]quinoline (XIIIa and b) and *dl*-8-acetyl-3,3a,4,5,8,9,10,10c-octahydro-4-(*p*-hydroxyphenyl)-10c-methyl-2*H*-furo[3,2-*c*]pyrrolo[3,2-*f*]quinoline (XIVa and b) (Chart III). Once again the condensation

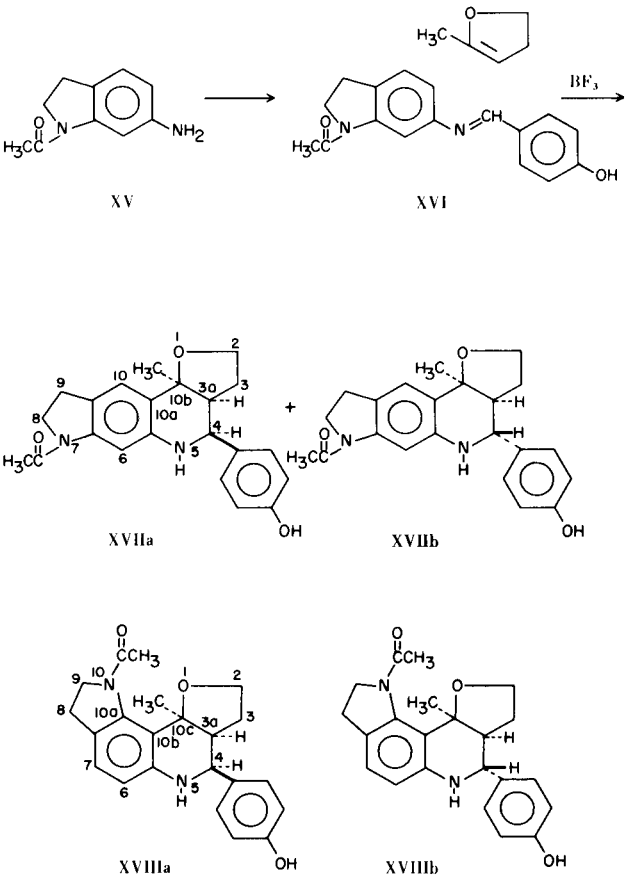
CHART III



proceeded slowly and was incomplete despite the addition of 2 portions of catalyst, excess 2,3-dihydro-5-methylfuran, and a prolonged reaction time. The product that was isolated and purified (48%) showed two overlapping spots on TLC (silica; ethyl acetate, triethylamine). The NMR spectrum of this material showed a 3 proton signal at 2.10 (acetyl-CH_3), 2.5 proton signal at 1.58 (angular-CH_3), a 0.5 proton signal at 1.24 (angular-CH_3), and a peak at 4.46 which integrated for nearly 1 proton. The peak at 4.46 clearly indicates that the 3a,4-protons of the predominant product in the mixture are in the *cis*-configuration. The aromatic region showed a 4 proton AB pattern from the *p*-hydroxyphenyl ring protons, and singlets at 6.53 and 7.97 integrating for approximately 1 proton each. Since these peaks do not indicate *ortho*-coupling as is required for the 6,7-protons of XIVa, structure XIIIa is tentatively assigned to the major component. The minor component with the peak at 1.24 is assigned structure XIIIb because of the correspondence of the chemical shift with IXb.

The condensation of 1-acetyl-6-[(*p*-hydroxybenzylidene)amino]indoline (XVI) with 2,3-dihydro-5-methylfuran (II) also required prolonged reaction time and the use of excess enol ether. Thin layer chromatography (silica; ethyl acetate, triethylamine) indicated that at least three of the expected reaction products, namely *dl*-7-

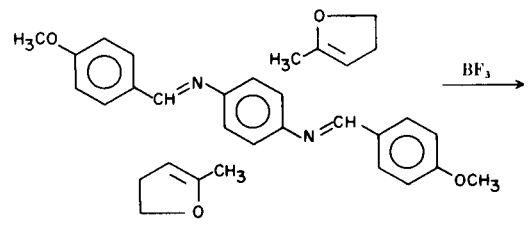
CHART IV



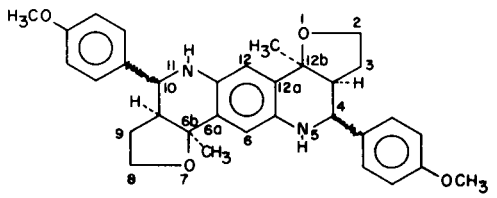
acetyl-3,3a,4,5,7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-*c*]pyrrolo[3,2-*g*]quinoline (XVIIa and b) and *dl*-10-acetyl-3,3a,4,5,8,9,10,10c-octahydro-4-(*p*-hydroxyphenyl)-10c-methyl-2*H*-furo[3,2-*c*]pyrrolo[2,3-*f*]quinoline (XVIIIa and b) (Chart IV), were present in the reaction mixture. However, the material that was isolated and purified (45%) showed only two spots on TLC. The NMR spectrum showed the acetyl peak at 2.10, and peaks of approximately equal intensity of 1.59 and 1.20. The peak at 4.46 had a relative integration for 0.4 protons. The compounds of this product are tentatively assigned structures XVIIa and b based on the upfield position of the 10b-CH_3 peak at 1.20. Structure XVIIIb would be expected to absorb in the 1.25 to 1.35 region. Further, the relative integration of the peak at 4.46 indicates a *cis-trans* mixture, since it is assumed that a signal from the *trans* 4-proton would be buried in other absorption in the 3.5-4.1 region.

The condensation of two equivalents of 2,3-dihydro-5-methylfuran (II) with one equivalent of *N,N'*-bis(*p*-methoxybenzylidene)-*p*-phenylenediamine in the presence of boron trifluoride gave a product, m.p. 305-308°, that analyzed correctly (C,H,N) for a double adduct $C_{32}H_{36}N_2O_4$. Unfortunately, the product was very insoluble in common solvents and a meaningful NMR curve could not be obtained. However, examination of Dreiding models of the two most likely products, namely *dl*-2,3,3a,4,5,6b,8,-9,9a,10,11,12b-dodecahydro-4,10-bis(*p*-methoxyphenyl)-6b,12b-dimethylfuro[3,2-*c*]furo[2',3':4,5]pyrido[2,3-g]-quinoline (XX) and *dl*-2,3,3a,4,5,8,9,9a,10,11,12a,12d-dodecahydro-4,9-bis(*p*-methoxyphenyl)-12a,12d-dimethyl-difuro[2,3-*a*:3',2'-*k*][4,7]phenanthroline (XXI) (Chart V), clearly indicates that XXI would be highly hindered in contrast to XX. Therefore structure XX is tentatively assigned.

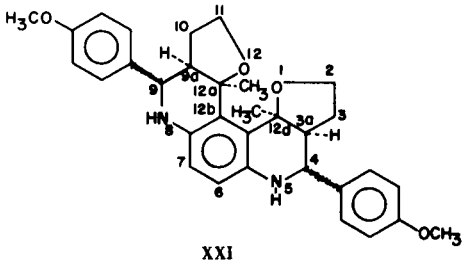
CHART V



XIX



XX



XXI

The intermediate Schiff bases 1-acetyl-1,2,3,4-tetrahydro-6-[(*p*-hydroxybenzylidene)amino]quinoline (VIII), 1-acetyl-5-[(*p*-hydroxybenzylidene)amino]indoline (XII), and 1-acetyl-6-[(*p*-hydroxybenzylidene)amino]indoline (XVI) were prepared from 6-nitro-1,2,3,4-tetrahydroquinoline (8), 5-nitroindoline, and 6-nitroindoline *via* the following general route. The nitro compound was acetylated using acetic anhydride in pyridine, and the resultant 1-acetyl-6-nitro-1,2,3,4-tetrahydroquinoline, 1-acetyl-5-nitroindoline, and 1-acetyl-6-nitroindoline were hydrogenated over Raney nickel to give 1-acetyl-6-amino-1,2,3,4-tetrahydroquinoline (VII), 1-acetyl-5-aminoindoline (XI) (9), and 1-acetyl-6-aminoindoline (XV) (10). Reaction of the respective amines with *p*-hydroxybenzaldehyde afforded the requisite Schiff bases.

EXPERIMENTAL (7)

Mixture of *dl*-10-Acetyl-2,3,3a,4,5,7,8,9,10,11b-decahydro-4-(*p*-hydroxyphenyl)-11b-methylfuro[3,2-*c*]pyrido[2,3-*g*]quinoline (IXa and b) and *dl*-8-acetyl-2,3,3a,4,5,8,9,10,11,11c-decahydro-4-(*p*-hydroxyphenyl)-11c-methylfuro[2,3-*a*][4,7]phenanthroline (Xa and b) (Chart II).

To a slurry of 2.94 g. (0.01 mole) of 1-acetyl-1,2,3,4-tetrahydro-6-[(*p*-hydroxybenzylidene)amino]quinoline (VIII) in ethyl acetate was added 2 drops of boron trifluoride etherate followed by 0.84 g. (0.01 mole) of 2,3-dihydro-5-methylfuran. After 2 hours 50 ml. of dioxane was added along with 2 drops of boron trifluoride etherate and 0.84 g. of 2,3-dihydro-5-methylfuran. After 3 hours, TLC (silica; ethyl acetate, triethylamine) showed no starting material. The mixture was filtered and the filtrate was concentrated to an oil. Crystallization from ethyl acetate gave 1.8 g. (48%) of yellow solid, m.p. 232-254°. The NMR spectrum showed peaks at 1.98, 1.58, 1.31 and 1.20.

Anal. Calcd. for $C_{23}H_{26}N_2O_3$: C, 72.95; H, 6.93; N, 7.40. Found: C, 72.82; H, 7.03; N, 7.27.

Mixture of *dl*-9-Acetyl-3,3a,4,5,7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-*c*]pyrrolo[2,3-*g*]quinoline (XIIIa and b) (Chart III).

To a slurry of 1-acetyl-5-[(*p*-hydroxybenzylidene)amino]indoline (XII) (5.60 g., 0.020 mole) in 200 ml. of ethyl acetate was

added 5 drops of boron trifluoride etherate followed by 1.68 g. (0.020 mole) of 2,3-dihydro-5-methylfuran. After 5 hours TLC (silica; ethyl acetate, triethylamine) indicated that little reaction had occurred. Dioxane (200 ml.) was added followed by an additional 5 drops of boron trifluoride etherate and 1.68 g. of 2,3-dihydro-5-methylfuran. After stirring at room temperature for an additional 18 hours, the reaction mixture was filtered and 2.9 g. (52%) of the Schiff base (XII) was recovered. TLC indicated that the filtrate contained at least two products. Concentration gave a syrup which upon crystallization from methanol-ethyl acetate yielded 0.4 g. of pale yellow crystals, m.p. 271-273°. Concentration of the filtrate and crystallization of the residue from *N,N*-dimethylformamide-water gave an additional 1.3 g. for a total of 1.7 g. (48% based on recovered starting material). The NMR spectra displayed peaks at 1.58 and 1.24.

Anal. Calcd. for $C_{22}H_{24}N_2O_3$: C, 72.50; H, 6.64; N, 7.69. Found: C, 72.27; H, 6.64; N, 7.66.

Mixture of *dl*-7-Acetyl-3,3a,4,5,7,8,9,10b-octahydro-4-(*p*-hydroxyphenyl)-10b-methyl-2*H*-furo[3,2-*c*]pyrrolo[3,2-*g*]quinoline (XVIIa and b) (Chart IV).

A slurry of 1-acetyl-6-[(*p*-hydroxybenzylidene)amino]indoline (XVI) (5.60 g., 0.020 mole) in 200 ml. of ethyl acetate was treated with 5 drops of boron trifluoride etherate and 1.68 g. (0.020 mole) of 2,3-dihydro-5-methylfuran. When, after 2 hours, TLC (silica; ethyl acetate, triethylamine) showed incomplete reaction, an additional 1.68 g. of 2,3-dihydro-5-methylfuran was added and the mixture was stirred an additional 18 hours, when TLC showed no starting material. The mixture was filtered, and the filtrate concentrated to obtain a second crop. There were combined and crystallized from methanol-ethyl acetate to give 3.3 g. (45%) of pale yellow crystals, m.p. 239-242°. The NMR spectrum gave peaks at 1.59 and 1.20.

Anal. Calcd. for $C_{22}H_{24}N_2O_3$: C, 72.50; H, 6.64; N, 7.69. Found: C, 72.45; H, 6.68; N, 7.86.

dl-2,3,3a,4,5,6b,8,9,9a,10,11,12b-Dodecahydro-4,10-bis(*p*-methoxyphenyl)-6b,12b-dimethylfuro[3,2-*c*]furo[2',3':4,5]pyrido[2,3-*g*]quinoline (XX) (Chart V).

To a slurry of 25.0 g. (0.075 mole) of *N,N'*-bis(*p*-methoxybenzylidene)-*p*-phenylenediamine (XIX) (11) in 500 ml. of benzene at 40° was added 0.5 ml. of boron trifluoride etherate followed by the dropwise addition of 12.2 g. (0.15 mole) of 2,3-dihydro-5-methylfuran in 500 ml. of benzene. After stirring for 3 hours an additional 0.25 ml. of boron trifluoride etherate and 6.1 g. of 2,3-dihydro-5-methylfuran were added in succession. This was repeated again after an additional 2 hours. After stirring overnight the product was collected by filtration and washed with dioxane to give 12.5 g. (32%), m.p. 305-308°. This compound was very insoluble in common solvents and a meaningful NMR curve could therefore not be obtained.

Anal. Calcd. for $C_{32}H_{36}N_2O_4$: C, 74.98; H, 7.08; N, 5.46. Found: C, 74.66; H, 7.07; N, 5.44.

1-Acetyl-1,2,3,4-tetrahydro-6-[(*p*-hydroxybenzylidene)amino]-quinoline (VIII).

A mixture of 9.35 g. (0.049 mole) of 1-acetyl-6-amino-1,2,3,4-tetrahydroquinoline (VII) and 5.98 g. (0.049 mole) of *p*-hydroxybenzaldehyde in 100 ml. of benzene was heated under reflux for 5 hours while 0.7 ml. of water was collected in a Dean-Stark trap. The reaction mixture was cooled and the precipitate collected. Recrystallization from acetonitrile gave 10.6 g. (71%), m.p. 186-188°.

Anal. Calcd. for $C_{18}H_{18}N_2O_2$: C, 73.45; H, 6.16; N, 9.52. Found: C, 73.54; H, 6.09; N, 9.43.

1-Acetyl-5-[(*p*-hydroxybenzylidene)amino]indoline (XII).

1-Acetyl-5-aminoindoline (XI) (9) (6.0 g., 0.034 mole) and *p*-hydroxybenzaldehyde (4.2 g., 0.034 mole) were combined in 200 ml. of 2-propanol and boiled under reflux for 4 hours. The resultant precipitate was collected and washed with 2-propanol to give 8.0 g. (84%), m.p. >250° (indefinite).

Anal. Calcd. for $C_{17}H_{16}N_2O_2$: C, 72.83; H, 5.75; N, 10.00. Found: C, 72.10; H, 5.79; N, 10.42.

1-Acetyl-6-[(*p*-hydroxybenzylidene)amino]indoline (XVI).

In like manner 8.8 g. of 1-acetyl-6-aminoindoline (XV) (10) was allowed to react with 6.1 g. of *p*-hydroxybenzaldehyde to give 11.5 g. (82%), m.p. 236-240°.

Anal. Calcd. for $C_{17}H_{16}N_2O_2$: C, 72.83: H, 5.75; N, 10.00. Found: C, 73.10; H, 6.05; N, 10.10.

1-Acetyl-6-amino-1,2,3,4-tetrahydroquinoline (VII).

A mixture of 10.8 g. (0.049 mole) of 1-acetyl-6-nitro-1,2,3,4-tetrahydroquinoline (8) and 1 g. of Raney-nickel catalyst in 50 ml. of methanol and 200 ml. of tetrahydrofuran was hydrogenated at an initial hydrogen pressure of 50 psig for 24 hours. The catalyst was removed by filtration, washed with *N,N*-dimethylformamide, and the filtrate evaporated to an oil which could not be crystallized. This crude product was used without further purification; infrared spectrum 1630 cm^{-1}.

Acknowledgments.

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