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Registry No. 1 (R_2 = H), 4562-27-0; 1 (R_2 = Me), 6104-45-6; 3 (R_1 = R_2 = H), 33612-61-2; 3 (R_1 = Me; R_2 = H), 70850-63-4; 3 (R_1 = H; R_2 = Me), 70850-64-5; 3 (R_1 = R_2 =Me) iodide, 14027-62-4; 3 (R_1 = R_2 = Me) perchlorate, 70850-65-6; 4 (R_2 = H), 19808-30-1; 4 (R_2 = Me), 14248-02-3; 5 (R_1 = Me), 70850-66-7; 6 (R_1 = R_2 = H) bromide, 70850-67-8; 6 (R_1 = Me; R_2 = H) bromide, 70850-68-9; 6 (R_1 = H; R_2 = Me), 70850-69-0; 6 (R_1 = R_2 = Me) perchlorate, 70850-71-4; 7 (R_1 = R_2 = H), 70850-72-5; 7 (R_1 = Me; R_2 = H), 70850-73-6; 7 (R_1 = H; R_2 = Me), 70850-73-6; 7 (R_1 = R_2 = Me), 70850-74-7; 16 (R_1 = R_2 = Me), 70850-75-8; 5-deuterio-1,4-dihydro-1,3-dimethyl-4-oxopyridinium iodide, 70850-76-9; 5-deuterio-1,4-dihydro-1,3-dimethyl-4-oxopyridinium perchlorate, 70850-78-1; 5-deuterio-3-methyl-4-pyrimidinone, 70850-79-2; 2-deuterio-3-methyl-4-pyrimidinone, 18542-92-2.

6,7-Dihydrocyclobuta[*g*]quinoline[1]

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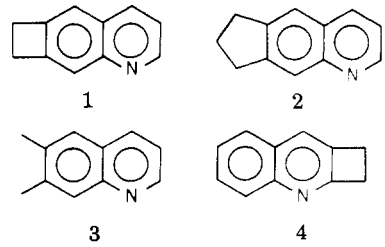
6,7-Dihydrocyclobuta[*g*]quinoline (**1**) was prepared by Skraup cyclization of 4-aminobenzocyclobutene. Similar reactions of 5-aminoindan and 3,4-dimethylaniline afforded as the major product 7,8-dihydro-6*H*-cyclopenta[*g*]quinoline (**2**) and 6,7-dimethylquinoline (**3**), respectively. Basicities of **1**–**3** and 1,2-dihydrocyclobuta[*b*]quinoline (**4**) were determined by potentiometric titration and the following pK_a values were deduced: **1**, 5.45; **2**, 5.54; **3**, 5.41; **4**, 3.99. The four-membered ring in **1** caused no reduction in basicity comparable to that observed with **4**. These data are consistent with orbital rehybridization induced by ring strain, an effect which is transmitted through the σ framework.

Twelve years ago the preparation and basicity of 1,2-dihydrocyclobuta[*b*]quinoline were reported, and it was established that the fusion of a small ring at the 2,3 position of an aza aromatic compound markedly decreased the basicity of such a strained heterocyclic system.[3] This effect, observed also with other quinoline,[4] quinoxaline,[5] and naphthyridine[6] systems, has been interpreted in terms of orbital hybridization concepts developed by Streitwieser and co-workers for strained carbocyclic compounds.[7] The smaller the fused cycloalkane moiety, the more pronounced is the effect on basicity. Although the initial report involved a fused four-membered ring,[3a] subsequent work demonstrated that five-membered rings exert a similar influence.[4-6] Heteroatoms can be incorporated into the strained ring, as shown by the extensive studies of derivatives of furo[2,3-*b*]pyridine and pyrano[2,3-*b*]pyridine.[8] Most recently, in an elegant series of papers by Thummel and Kohli, the same correlation between increased ring strain and decreased basicity was observed in a variety of mono- and bisannelated pyridines.[9-11]

The utility of the heteroaromatic systems lies in the lone pair of electrons on nitrogen which serves (under equilibrium conditions of potentiometric titration) as a probe for the strain introduced by the adjacent ring. In all of the cases cited above the nonbonded pair of electrons on N occupies a nominal sp^2 hybrid orbital which, as a result of increased s character, renders the electron pair less available as a Brønsted base. The nonbonding pair, therefore, is isoelectronic with the aryl carbanions generated by base-catalyzed proton abstraction from carbon-1 of biphenylene[7] and carbon-3 of benzocyclobutene.[12] Orbital rehybridization was postulated to account for the increased kinetic acidity of aryl hydrogens at sites adjacent to the junction of strained rings. Since this interpretation posits a redistribution of the fraction of s and p character in the three hybrid orbitals of a bridgehead carbon, the impact of such rehybridization is transmitted by the σ framework of a molecule. As a consequence, the effect should be attenuated by an increase in the number of σ bonds between the strained ring junction and the site of reactivity. Such a relationship was observed between positions 1 and 2 of biphenylene.[7] At the inception of the present work there had been no report of the dependence of heterocyclic basicity on the position of a strained ring. The recent work on cyclobuta[*b*]- and cyclobuta[*c*]-pyridines has provided the first example in a heterocyclic system.[10]

The compounds chosen for our study were 6,7-dihydrocyclobuta[*g*]quinoline (**1**), 7,8-dihydro-6*H*-cyclopenta[*g*]quinoline (**2**), and 6,7-dimethylquinoline (**3**). The



1 **2**

3 **4**

series was suitable for basicity comparisons with the previously reported 1,2-dihydrocyclobuta[*b*]quinoline (**4**).[3]

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Table I. Basicities of Selected Quinolines

compd	HNP,[a] mV	pK_a	ref
1	351	5.45	*b*
2	346	5.54	*b*
3	353.5	5.41	*b*
4	431	3.99	*b*
2-methylquinoline	340	5.70	19
4-methylquinoline	355	5.33	20
isoquinoline	356	5.33	21
quinoline	375	5.06	19
6-methylquinoline	377	4.99	22

[a] Average values (±1 mV) from duplicate titrations. [b] Present work.

Compounds 1–3 were prepared by Skraup cyclizations of the appropriate aryl amines: 4-aminobenzocyclobutene (**5**), 5-aminoindan (**6**), and 3,4-dimethylaniline (**7**), respectively. In each case the cyclization could afford a mixture of the desired 6,7-disubstituted quinoline (1–3) and the isomeric 5,6-disubstituted quinoline. The product mixture obtained from **5** only contained 1 (75%) and dimethylquinolines. The absence of 7,8-dihydrocyclobuta[*f*]quinoline was in accord with our expectations, since the key step in the Skraup synthesis is an electrophilic aromatic substitution and the benzocyclobutene system is known to undergo S_E reactions at positions β to the four-membered ring.[13] The conversions of **6** and **7** to quinolines are known to give isomeric products. From **6** was obtained a 95:5 mixture of **2** and 8,9-dihydro-7*H*-cyclopenta[*f*]quinoline (**8**),[14] while **7** afforded a ca. 70:30 mixture of **3** and 5,6-dimethylquinoline (**9**).[15] The ratio of **2**:**8** (compared to **3**:**9**) is another example of the reduced attack of electrophiles at positions α to fused, strained rings. Studies with indan were the first report of such relative reactivity and the resulting explanation became known as the Mills–Nixon effect.[16] This interpretation, proposed 50 years ago, invokes bond fixation in strained arenes and it continues to provoke active research.[17]

Compound 1 has not previously been reported. The assigned structure was consistent with its mode of preparation, elemental analyses, high-resolution mass spectrum, and proton NMR spectrum. The latter data, in particular, confirmed the structure as the dihydrocyclobuta[*g*] derivative. At 100 MHz the methylene protons appeared as a singlet, but at 270 MHz their nonequivalence was evidenced by two peaks separated by 0.009 Hz. The assignment of the individual aryl protons, especially the H-5 and H-8 singlets, precluded the possibility of a dihydrocyclobuta[*f*] structure which would exhibit a large J_{78}.[18] The $J(^{13}C-H)$ value of the methylene protons was 133 Hz, somewhat less than that observed for **4**.[3b]

Basicities of 1–4, determined as half-neutralization potentials (HNP) by titration at 25 °C in acetic anhydride with 0.10 N perchloric acid in acetic acid, are listed in Table I. The apparent acid dissociation constants (pK_a) were derived graphically from a plot of HNP vs. known pK_a's for a series of quinolines. The assumption that HNP (Ac_2O) and pK_a (H_2O) were linearly related, although known for some time,[19,23] has been questioned.[24] Therefore, the data used in the present work to verify the linear relationship are included in Table I. From the basicity measurements it was clear that the strained ring in **1** exerted no special effect compared to **2** and **3**. Thus, the results were consistent with orbital rehybridization. Comparable data were reported quite recently for the isomeric cyclobuta- and cyclopentapyridines.[10] In the present study compounds 1–3 were slightly more basic than either 6-methylquinoline[20,22,25] or 7-methylquinoline,[20,25,26] but the differences were probably not significant.

Experimental Section

Boiling points and melting points are uncorrected; the latter were determined on a modified Hershberg apparatus with total-immersion Anschutz thermometers. Spectra were recorded on the following instruments: a Perkin-Elmer 237B IR spectrophotometer; a Cary 14 UV spectrophotometer; an AEI MS-9 mass spectrometer at an ionizing voltage of 70 eV; Perkin-Elmer R12B and Varian HR-100 (equipped with field frequency lock) NMR spectrometers, and the 270 MHz instrument (A. H. Redfield modification) at Brandeis University. Chemical shifts are reported in parts per million from an internal Me_4Si reference; coupling constants (J) are expressed in hertz; signal multiplicity: s = singlet, dd = doublet of doublets, t = triplet, m = multiplet. Analytical gas–liquid chromatography (GLC) was performed on a Varian Aerograph 1400 instrument equipped with a 12 ft × $^1/_8$ in. column packed with 10% Dow-Corning 710 on Chromosorb W with helium as the carrier gas. Preparative GLC was performed on a Wilkens Aerograph A-90-P instrument fitted with a 20 ft × $^3/_8$ in. column packed with 20% Dow-Corning 710 on Chromosorb G with helium as the carrier gas. Column temperature and helium flow rate are described in parentheses; retention times of peaks are reported in minutes. Microanalyses were performed by Galbraith Laboratories, Inc., Knoxville, Tenn.

Materials. Benzocyclobutene was prepared by known methods,[27,28] bp 147–150 °C. Commercially available samples of quinoline, isoquinoline, 2-methylquinoline, 4-methylquinoline, and 6-methylquinoline were vacuum distilled immediately prior to basicity measurements.

4-Nitrobenzocyclobutene. Nitration of benzocyclobutene at ice temperature in acetic anhydride–acetic acid[29] afforded 4-nitrobenzocyclobutene in 39% yield: bp 98.5–105 °C (0.8 mm) [lit.[30] bp 70–85 °C (0.1 mm)]; GLC (215 °C, 28 mL/min) 4-nitro-*o*-xylene 5.9 (trace), 4-nitrobenzocyclobutene 7.2; IR (CCl_4) same as literature values;[29] NMR (60 MHz, CCl_4) δ 2.37 (s, 4), 7.1–8.2 (m, 3).

4-Aminobenzocyclobutene (5). Catalytic hydrogenation of 4-nitrobenzocyclobutene in absolute EtOH over platinum oxide catalyst afforded **5** in quantitative yield as an orange oil, which was used without further purification:[30] GLC (215 °C, 30 mL/min) **7** 3.3 (trace), **5** 4.3; NMR (60 MHz, CCl_4) δ 3.01 (s, 4), 3.72 (s, 2), 6.3–7.1 (m, 3).

6,7-Dihydrocyclobuta[*g*]quinoline (1). To a magnetically stirred mixture of **5** (44.8 g, 0.376 mol), ferrous sulfate heptahydrate (12.3 g, 0.044 mol), and nitrobenzene (35.8 g, 0.291 mol) was added a solution of boric acid (21.6 g, 0.349 mol) in glycerol

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(135.6 g, 147 mol). To the ice-cold, stirred mixture was added concentrated sulfuric acid (64 mL) in small portions. The reaction mixture was refluxed for 3 h, basified with 25% NaOH (575 mL), diluted with water, and steam-distilled and the distillate (7 L) was extracted with benzene. The combined benzene extract was dried (Na_2SO_4), refluxed for 30 min with *p*-toluenesulfonyl chloride (15.3 g, 0.080 mol), poured into ice water (300 g), and acidified (Congo Red end point) with concentrated HCl. The aqueous phase was washed with benzene, basified with 6 N NaOH to pH 10, and extracted with CH_2Cl_2. The combined CH_2Cl_2 extract was dried (Na_2SO_4), distilled to remove solvent, and vacuum distilled to remove quinoline (head temperature 128 °C at 1.4 mm). The distillate (6.99 g) solidified to orange-brown crystals: GLC (215 °C, 30 mL/min) **3** 9.5, **9** 10.2, **1** 12. Integration of the peak areas indicated 23% of dimethylquinolines (**3** + **9**) and 75% of **1** (5.2 g, 0.034 mol, 9.0%) which was purified and isolated by preparative GLC [(232 °C, 200 mL/min) quinoline 39, **3** 46, **9** 48.5, **1** 67] to give **1**: mp 78.6–79.6 °C; UV_{max} (95% EtOH) 291 nm (ϵ 3810), 297.5 (3740), 303.5 (5110), 310.5 (4680), 317.5 (8220); NMR (100 MHz, $DCCl_3$) δ 3.40 (s, 4), 7.44 (dd, J_{23} = 4, J_{34} = 8 Hz, H-3), 7.51 (s, H-5), 7.86 (s, H-8), 8.25 (dd, J_{24} = 2, J_{34} = 8 Hz, H-4), 8.86 (dd, J_{23} = 4, J_{24} = 2 Hz, H-2); at 270 MHz ($DCCl_3$) the methylene protons were separated by 0.0093 ppm; high-resolution MS *m/e* 155.07346 (calcd for $C_{11}H_9N$, 155.07354) (M^+).

Anal. Calcd for $C_{11}H_9N$: C, 85.13; H, 5.85; N, 9.02. Found: C, 85.21; H, 5.75; N, 9.11.

7,8-Dihydro-6*H*-cyclopenta[*g*]quinoline (2). A Skraup cyclization was carried out in a manner identical with that reported above, using the following conditions: **6** (8.5 g, 64 mmol), ferrous sulfate heptahydrate (2.5 g, 9.0 mmol), 5-nitroindan (7.5 g, 46 mmol), boric acid (4.4 g, 71 mmol), glycerol (28 g, 0.30 mol), concentrated H_2SO_4 (13 mL); reflux time, 4 h. The yellow solid (0.84 g) obtained as a crude product was chromatographed on a column of alumina (Woelm, neutral) with $HCCl_3$ as eluant to give 0.26 g (2.4%) of white crystalline **2**: mp 77.4–78.0 °C (lit.[14] mp 79–80.5 °C); GLC (220 °C, 29 mL/min) **2** 16.2, no peaks corresponding to **6** (5.9) or 5-nitroindan (9.9); IR ($HCCl_3$) 2950, 2850, 870 cm^{-1}; UV_{max} (95% EtOH) 307 nm (ϵ 6010), 310 (5000), 320 (9000); NMR (60 MHz, $DCCl_3$) δ 2.25 (m, 2), 3.12 (t, 4), 7.18–8.97 (m, 5); high-resolution MS *m/e* 169.08936 (calcd for $C_{12}H_{11}N$, 169.08920) (M^+); picrate mp 274–275 °C dec (95% EtOH) (lit.[14] mp 269–271 °C).

Subsequent fractions from the column chromatography afforded a small amount of yellow oil, considered to be **8**: GLC (220 °C, 29 mL/min) **8** 18; NMR (60 MHz, $DCCl_3$) δ 2.25 (m, 2), 3.16 (m, 4), 7.15–8.95 (m, 5) (lit.[14] mp 43–44.5 °C, picrate mp 190–191 °C).

6,7-Dimethylquinoline (3). A Skraup cyclization was carried out in a manner identical with that reported above, using the following conditions: **7** (60.0 g, 0.495 mol), ferrous sulfate heptahydrate (16.2 g, 0.058 mol), 4-nitro-*o*-xylene (47.9 g, 0.317 mol), boric acid (28.5 g, 0.461 mol), glycerol (179 g, 1.94 mol), concentrated H_2SO_4 (84 mL); reflux time, 4 h. The red-brown residual oil (65.7 g) obtained as a crude product was vacuum distilled to give 57.8 g (74.3%) of 6,7-dimethylquinoline (**3**) and 5,6-dimethylquinoline (**9**): bp 100–107 °C (1.8 mm) [lit.[15a] bp 152–160 °C (15 mm)]; GLC (215 °C, 30 mL/min) **3** 10.2, **9** 11. Separation of the product mixture by preparative GLC [(235 °C, 200 mL/min) **3** 56, **9** 59] gave white crystalline **3**: NMR (60 MHz, CCl_4) δ 2.41 (s, 3), 2.44 (s, 3), 7.1–8.1 (m, 4), 8.8 (m, 1, H-2). Obtained similarly was **9**: NMR (60 MHz, CCl_4) δ 2.47 (s, 3), 2.53 (s, 3), 7.1–8.5 (m, 4), 8.8 (m, 1, H-2).

Basicity Measurements. Basicities were determined at 25.00 ± 0.02 °C by our previously reported procedure,[5] using a Beckman Century SS-1 pH meter. The end point and half-neutralization potential were determined graphically. All runs were carried out in duplicate, with a precision of ±1 mV.

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Registry No. **1**, 70891-81-5; **2**, 7193-31-9; **3**, 20668-33-1; **4**, 13353-49-6; **5**, 55716-66-0; **6**, 24425-40-9; **7**, 95-64-7; **8**, 70891-82-6; **9**, 20668-30-8; benzocyclobutene, 694-87-1; 4-nitrobenzocyclobutene, 52961-81-6; 4-nitro-*o*-xylene, 99-51-4.

Notes

Volume Profile of the Degenerate Equilibration of a Classical Norbornyl Ion

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In 1965, we reported that the hydrolysis of *exo*-2-norbornyl tosylate in aqueous acetone is characterized by an activation volume about 3–4 cm^3/mol less negative than that of the endo epimer and that the latter reaction is closely similar in its response to pressure to that of cyclopentyl tosylate.[2] Some time later we published a similar difference between the hydrolyses of the epimeric 3-substituted bicyclo[3.1.0]hexanes,[3] this time with the endo derivative less sensitive to pressure. These results are in agreement with the concept of σ participation, and we interpreted them in terms of the controversial[4] nonclassical ions, in which the positive charge is equally divided between two or more carbon atoms because of symmetry. Reduced volume contractions were subsequently observed in various solvolytic reactions involving phenyl participation as well.[5]

The argument is based on the Drude–Nernst equation,

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