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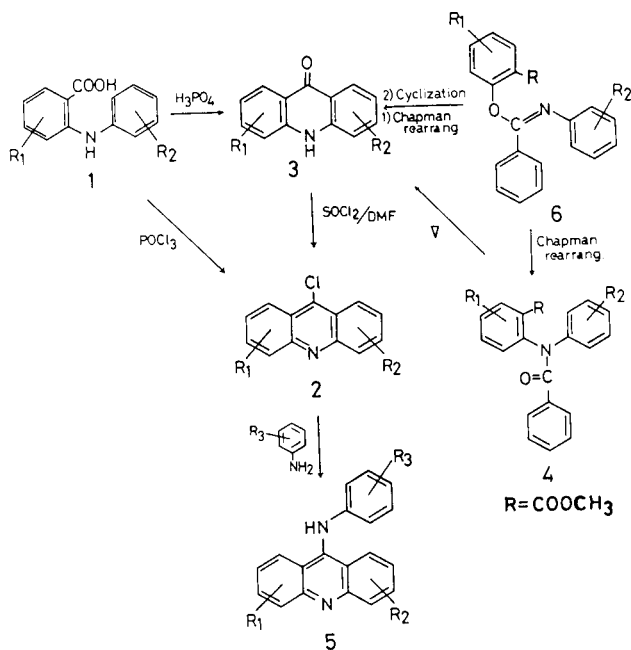
Dibenz[*c,h*]acridine (**13**) was formed directly by the Chapman rearrangement of 2-methoxycarbonyl-1-naphthyl-*N*-1'-naphthylbenzimidate (**11**) without isolation of intermediate **12**. When the Chapman rearrangement was carried out under mild reaction conditions, the intermediate **12** was isolated in high yield, whose structure was determined by the X-ray studies. The mechanism of the Chapman rearrangement can be interpreted on the basis of the X-ray data of **11** and **12**.

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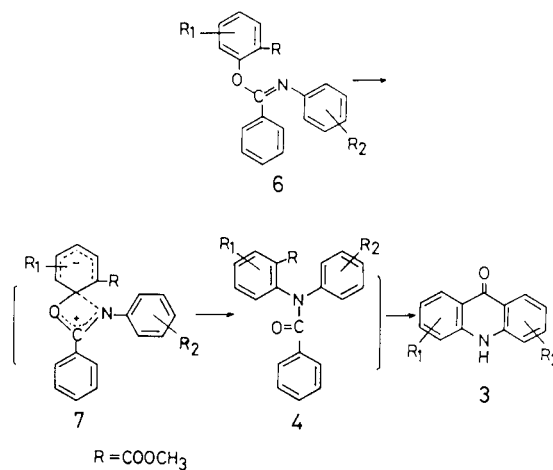
Since the acridines have been found as a high-boiling fraction in coal tar, they have been the subject of much technical and scientific interest. They have also provided a large number of basic dyestuffs, such as orange, yellow red, and purple dyes. In addition, acridine derivatives have provided useful chemotherapeutic drugs which have antibacterial, antimalarial, mutagenic and carcinogenic properties [2-6]. Recently, Cain *et al.* have synthesized a large number of 9-anilinoacridines and tested the anti-tumor activity employing the intraperitoneally (ip) implanted L1210 leukemia in mice [7-14]. Some of their 9-anilinoacridines, particularly the 4'-(9-acridinylamino)-

methanesulfonanilide (AMSA) [15-19] have been shown to have a broad spectrum action against a number of animal tumor system, which are also being evaluated clinically in the treatment of a number of human tumors.

Regarding the preparation of these 9-anilinoacridines **5**, arylamino side chains were coupled with 9-chloroacridines **2** which were obtained directly from the ring closure of an *N*-arylanthranilic acid (**1**) with phosphoryl chloride [20-22]. But the ring closure with phosphoryl chloride was not satisfactory, this was effected with either sulfuric acid, phosphoric acid, PPA and PPE to give the 9-(10*H*)-acridinones **3**, which were easily converted to the desired 9-chloroacridines **2** with thionyl chloride/DMF [23]. One of the important methods of preparation of these 9-(10*H*)-acridinones **3**, which were easily converted to the desired 9-chloroacridines **2** with thionyl chloride/DMF [23]. One of the important methods of preparation of these 9-(10*H*)-acridinones is the utilization of the Chapman rearrangement [24,25] of *N*-arylbenzimidates **6** shown in Scheme 1. The usefulness of the Chapman rearrangement is that it has been carried



Scheme 1. The substituents R₁, R₂, and R₃ are alkyl, alkoxy, alkoxy halogen or nitrogen groups.



Scheme 2. The substituents R₁, R₂, and R₃ are alkyl, alkoxy, alkoxy halogen or nitrogen groups.

Table 2

Final Positional and Thermal Parameters of Non-hydrogen and Hydrogen Atoms with Estimated Standard Deviations

$$B_{eq} = \frac{1}{3} (U_{11} + U_{22} + U_{33}) \text{ (where } U_{11} = \frac{B_{11}}{8\pi^2 a^*{}^2}, \text{ ect.)}$$

Atoms	x	y	z	Beq or B/Å ²
N 1	0.1665(3)	0.4565(4)	0.5936(3)	3.31
C 2	0.1695(4)	0.3447(5)	0.5885(3)	3.57
C 3	0.2296(4)	0.3004(6)	0.5849(4)	4.84
C 4	0.2352(5)	0.1903(6)	0.5778(4)	5.90
C 5	0.1786(4)	0.1297(5)	0.5754(3)	5.77
C 6	0.1148(4)	0.1736(6)	0.5789(4)	4.75
C 7	0.0546(3)	0.1093(4)	0.5769(3)	6.12
C 8	-0.0063(3)	0.1520(4)	0.5813(3)	6.83
C 9	-0.0121(4)	0.2600(7)	0.5867(4)	6.26
C 10	0.0445(4)	0.3244(6)	0.5884(4)	4.67
C 11	0.1097(4)	0.2836(5)	0.5857(3)	3.88
C 12	0.1964(3)	0.5156(5)	0.5493(3)	3.42
C 13	0.2589(4)	0.5713(5)	0.5743(3)	3.77
C 14	0.2847(4)	0.6260(6)	0.5275(4)	4.60
C 15	0.2520(4)	0.6222(6)	0.4582(4)	4.90
C 16	0.1885(4)	0.5655(5)	0.4303(3)	4.01
C 17	0.1522(4)	0.5594(6)	0.3573(4)	5.33
C 18	0.0898(4)	0.5072(6)	0.3311(4)	6.01
C 19	0.0596(4)	0.4592(7)	0.3758(4)	5.41
C 20	0.0930(4)	0.4613(6)	0.4470(4)	4.22
C 21	0.1583(3)	0.5129(5)	0.4763(3)	3.48
C 22	0.3023(4)	0.5731(6)	0.6504(4)	4.58
O 23	0.3060(5)	0.5037(7)	0.6909(4)	5.53
O 24	0.3356(4)	0.6635(7)	0.6665(4)	5.96
C 25	0.3812(5)	0.6768(7)	0.7386(5)	9.07
C 26	0.1578(4)	0.4979(5)	0.6538(3)	4.02
O 27	0.1543(3)	0.4425(4)	0.7003(3)	5.63
C 28	0.1496(4)	0.6141(5)	0.6554(4)	4.17
C 29	0.1072(4)	0.6721(6)	0.5997(4)	4.51
C 30	0.1001(4)	0.7792(6)	0.6069(4)	5.53
C 31	0.1366(6)	0.8283(9)	0.6680(5)	6.71
C 32	0.1788(5)	0.7734(6)	0.7235(4)	6.85
C 33	0.1847(5)	0.6653(6)	0.7187(4)	5.85
H 3	0.268(1)	0.344(1)	0.588(1)	3.43
H 4	0.282(4)	0.156(6)	0.578(4)	3.47
H 5	0.179(4)	0.058(6)	0.569(4)	3.13
H 7	0.057(4)	0.028(6)	0.573(4)	5.16
H 8	-0.052(5)	0.104(7)	0.594(5)	4.34

H 9	-0.051(4)	0.284(7)	0.604(4)	4.11
H 10	0.041(4)	0.392(7)	0.591(4)	3.01
H 14	0.326(3)	0.664(5)	0.542(3)	3.20
H 15	0.275(4)	0.657(6)	0.426(4)	3.79
H 17	0.177(4)	0.588(6)	0.327(4)	4.25
H 18	0.044(4)	0.498(7)	0.280(4)	4.50
H 18	0.044(4)	0.498(7)	0.280(4)	4.50
H 19	0.035(4)	0.411(7)	0.342(4)	5.46
H 20	0.072(5)	0.427(7)	0.476(5)	2.68
1H25	0.422(5)	0.690(8)	0.793(5)	5.13
2H25	0.354(9)	0.672(11)	0.763(10)	6.58
3H25	0.390(5)	0.728(8)	0.705(5)	6.39
H 29	0.076(4)	0.635(6)	0.553(4)	2.15
H 30	0.069(4)	0.822(6)	0.571(4)	2.65
H 31	0.136(4)	0.903(6)	0.673(4)	2.30
H 32	0.210(4)	0.825(6)	0.765(4)	5.94
H 33	0.208(5)	0.624(7)	0.758(5)	6.14

substituted compounds react more rapidly than the R, *para* compounds. That is, the three points; 1) the combination of resonance, 2) inductive electron-attracting effects, and 3) steric acceleration due to hindered rotation effects, play an important role in the rearrangement. These facts led us to find the existence of non-bonded N...C interaction in the N-arylbenzimidates [1] on the basis of three dimensional structural information of **11** obtained by X-ray analysis (Figure 2). When the Chapman rearrangement of **11** was carried out under mild reaction condition (240-250°), the intermediate **12** was isolated in high yield as shown in Scheme 3. Dibenz[*c,h*]acridine **13** was also formed by heating **12** to 300°. These interesting facts encouraged us to elucidate the three dimensional structure of **12** by means of X-ray to interpret the reaction mechanism in more detail.

Results and Discussion.

The established molecular structure and the labeling sequence of **12** is presented in the ORTEP drawing of Figure 1 [37]. Bond lengths and angles are shown in Table 3. The average C-C lengths of two naphthalene rings A and B are 1.397 and 1.397 Å, respectively (compared with 1.399 and 1.399 Å of **11**), while that of the phenyl ring C is 1.388 Å (1.390 Å for **11**). The methoxycarbonyl group has bond lengths of 1.460(12), 1.324(11), and 1.200(10) Å in comparison with 1.440(5), 1.341(5), and 1.205(5) Å in **11**, respectively. With respect to N1 atom, both the bond lengths of N1-C12, and N1-C2 are 1.443 Å. These values are normal, and especially, the bond lengths of two naphthalenes (ring A and B) are exactly equivalent within experimental error. That is, the normal bond lengths and angles of **12** show that compound **12** which resulted from

Table 3
Bond Lengths and Angles

a) Bond lengths for non-hydrogen atoms

N 1 - C 2	1.443(8)	N 1 - C 12	1.443(7)
N 1 - C 26	1.396(8)	C 2 - C 3	1.348(9)
C 2 - C 11	1.411(9)	C 3 - C 4	1.431(10)
C 4 - C 5	1.356(10)	C 5 - C 6	1.413(10)
C 6 - C 7	1.444(9)	C 6 - C 11	1.428(9)
C 7 - C 8	1.359(7)	C 8 - C 9	1.400(10)
C 9 - C 10	1.388(10)	C 10 - C 11	1.416(9)
C 12 - C 13	1.381(8)	C 12 - C 21	1.430(8)
C 13 - C 14	1.408(9)	C 13 - C 22	1.506(9)
C 14 - C 15	1.346(10)	C 15 - C 16	1.409(9)
C 16 - C 17	1.424(9)	C 16 - C 21	1.434(8)
C 17 - C 18	1.360(10)	C 18 - C 19	1.386(10)
C 19 - C 20	1.381(9)	C 20 - C 21	1.408(9)
C 22 - O 23	1.200(10)	C 22 - O 24	1.324(11)
O 24 - C 25	1.460(12)	C 26 - O 27	1.202(8)
C 26 - C 28	1.504(9)	C 28 - C 29	1.389(9)
C 28 - C 33	1.410(10)	C 29 - C 30	1.395(10)
C 30 - C 31	1.372(12)	C 31 - C 32	1.362(12)
C 32 - C 33	1.401(11)		

b) Bond lengths involving hydrogen atoms

H 3 - C 3	0.94(1)	H 4 - C 4	1.02(7)
H 5 - C 5	0.93(8)	H 7 - C 7	1.05(7)
H 8 - C 8	1.19(8)	H 9 - C 9	0.99(7)
H 10 - C 10	0.87(8)	H 14 - C 14	0.91(6)
H 15 - C 15	1.02(7)	H 17 - C 17	0.97(6)
H 18 - C 18	1.13(8)	H 19 - C 19	0.93(8)
H 20 - C 20	0.94(8)	1H25 - C 25	1.16(9)
2H25 - C 25	0.85(18)	3H25 - C 25	1.00(9)
H 29 - C 29	1.06(7)	H 30 - C 30	0.97(7)
H 31 - C 31	0.97(7)	H 32 - C 32	1.10(7)
H 33 - C 33	0.94(9)		

c) Bond angles

C 12 - N 1 - C 2	116.4(5)	C 26 - N 1 - C 2	117.4(5)
C 26 - N 1 - C 12	122.7(5)	C 11 - C 2 - C 3	121.0(6)
C 11 - C 2 - N 1	120.3(5)	C 3 - C 2 - N 1	118.8(5)
C 4 - C 3 - C 2	121.5(6)	C 5 - C 4 - C 3	118.7(7)
C 6 - C 5 - C 4	121.2(6)	C 11 - C 6 - C 7	119.0(6)
C 11 - C 6 - C 5	119.6(6)	C 7 - C 6 - C 5	121.4(6)
C 8 - C 7 - C 6	121.0(5)	C 9 - C 8 - C 7	120.2(5)
C 10 - C 9 - C 8	120.4(6)	C 11 - C 10 - C 9	121.5(6)
C 10 - C 11 - C 6	117.7(6)	C 10 - C 11 - C 2	124.2(6)
C 6 - C 11 - C 2	118.0(6)	C 21 - C 12 - C 13	120.2(5)

C 21 - C 12 - N 1	116.6(5)	C 13 - C 12 - N 1	123.2(5)
C 22 - C 13 - C 14	118.2(6)	C 22 - C 13 - C 12	122.2(5)
C 14 - C 13 - C 12	119.6(6)	C 15 - C 14 - C 13	122.0(6)
C 16 - C 15 - C 14	120.3(6)	C 21 - C 16 - C 17	118.4(6)
C 21 - C 16 - C 15	119.5(6)	C 17 - C 16 - C 15	122.1(6)
C 18 - C 17 - C 16	121.6(6)	C 19 - C 18 - C 17	119.8(6)
C 20 - C 19 - C 18	121.1(6)	C 21 - C 20 - C 19	121.0(6)
C 20 - C 21 - C 16	118.1(5)	C 20 - C 21 - C 12	123.7(5)
C 16 - C 21 - C 12	118.2(5)	O 24 - C 22 - O 23	124.6(8)
O 24 - C 22 - C 13	110.0(6)	O 23 - C 22 - C 13	125.5(7)
C 25 - O 24 - C 22	116.6(7)	C 28 - C 26 - O 27	122.5(6)
C 28 - C 26 - N 1	116.2(5)	O 27 - C 26 - N 1	121.2(6)
C 33 - C 28 - C 29	118.9(6)	C 33 - C 28 - C 26	117.2(6)
C 29 - C 28 - C 26	123.8(6)	C 30 - C 29 - C 28	119.9(7)
C 31 - C 30 - C 29	120.4(7)	C 32 - C 31 - C 30	120.9(9)
C 33 - C 32 - C 31	119.9(7)	C 32 - C 33 - C 28	119.9(7)

Table 4

Least-Squares Planes and Deviations (d/Å) of Atoms from the Plane

Plane A: Defined by the atoms C2 - C11

$$-0.01826X + 0.08409Y - 0.99629Z + 10.87103 = 0^*$$

C 2	-0.0092;	C 3	-0.0102;	C 4	0.0040
C 5	0.0049;	C 6	0.0097;	C 7	0.0001
C 8	-0.0168;	C 9	-0.0005;	C 10	0.0173
C 11	0.0007				

Plane B: Defined by the atoms C12 - C21

$$0.56032X - 0.82568Y - 0.06548Z + 6.04043 = 0$$

C 12	0.0375;	C 13	0.0196;	C 14	-0.0403
C 15	-0.0219;	C 16	0.0099;	C 17	0.0325
C 18	0.0207;	C 19	-0.0287;	C 20	-0.0319
C 21	0.0035				

Plane C: Defined by the atoms C28 - C33

$$0.93770X + 0.14320Y - 0.31657Z + 4.13403 = 0$$

C 28	0.0114;	C 29	0.0106;	C 30	-0.0128
C 31	0.0077;	C 32	0.0127;	C 33	-0.0168

Dihedral Angles between Planes

A and B 90.82 °

A and C 72.93

B and C 64.67

* Expressed in an orthogonal coordinate system in Å unit.

the Chapman rearrangement has the normal molecular geometry. In Table 4, the best least-squares planes are given together with the displacement of the atoms from the best planes. The dihedral angle between the planes of the two naphthalene rings A, and B is 90.82° (compared with 106.91° for **11**), while the dihedral angles between the phenyl ring C and the naphthalene rings A, and B are 71.93° and 64.67° which are comparable to 110.95° and 70.36° in **11**, respectively. On the basis of the molecular geometry described above, the accurate molecular conformation of compound **12** can be elucidated.

Regarding the mechanism of the Chapman rearrangement, it has been shown to be an intramolecular reaction in which a 1,3-shift of an aryl group from oxygen to nitrogen takes place. That is, the reaction requires the formation of a four-membered ring in the transition state and may be considered as a nucleophilic attack by nitrogen on the migrating aryl group. These facts could be interpreted by the existence of the non-bonded N...C interaction (2.648 \AA) [1] found in the linkage of the N17 and C1 atoms in **11** much less than the sum of the van der Waals radii (about 3.5 \AA) [26] (Figure 2). But, in the intermediate **12**, there are not the short interatomic distances that exist in **11**. By heating at 300° , the elimination of a methanol molecule occurs in compound **12** to give the dibenz[*c,h*]acridine **13**. It appeared to be natural that the dibenz[*c,h*]acridine **13** was formed in higher yield from **11** than **12**.

There are no unusually close intermolecular contacts, the shortest being carbon-carbon of 3.204 \AA which is greater than the sum of the van der Waals radii. All other intermolecular contacts correspond to normal van der Waals interactions.

EXPERIMENTAL

Compound **11** was prepared as described earlier (Scheme 3) [32]. Compound **11** was heated to 250° , whereupon a reaction set in to give **12**. The crystals of **12** used for the X-ray investigations reported were grown by a slow evaporation of a pyridine solution at room temperature. The nmr, ir and mass spectra supported the structure of **12**, mp $165\text{--}167^\circ$; ms: m/z 431 (M^+ , 100% relative intensity), 400 ($M^+ \cdot \text{CH}_3\text{O}$, 15); ir (Nujol): $1710, 1670 \text{ cm}^{-1}$; ^1H nmr (deuteriochloroform): δ 3.80 (3H, s, CH_3), and 6.90-8.65 (18H, m, aromatic protons).

Collection and Reduction of Data.

The crystal selected for the measurement of lattice parameters and intensity data was a cleaved fragment which had approximate dimensions of $0.30 \times 0.35 \times 0.30 \text{ mm}$. A preliminary examination of the crystal carried out on Rigaku automated, four-circle diffractometer showed it to be monoclinic. Systematic absences of $h + k = 2n + 1$, and $l = 2n + 1$ for $h0l$ established the space group to belong to $C2/c$. The unit cell parameters at room temperature were refined by least-squares using the Bragg angles ($\text{CuK}\alpha$, $\lambda = 1.54178 \text{ \AA}$) of 24 reflections ($45^\circ < 2\theta < 55^\circ$). The unit cell dimensions and crystal data are listed in Table 1. Intensity data were collected at room temperature on a diffractometer utilizing nickel-filtered $\text{CuK}\alpha$ radiation. The θ - 2θ scan mode was employed. The scan rate was 4° min^{-1} and the scan range in θ was varied by $1.2^\circ + 0.15^\circ \tan\theta$. Backgrounds were counted for 5s at both ends of the scan

with an offset of 50% of the scan range from the calculated position of the $\text{K}\alpha$ peak. A total of 3819 unique reflections of the type $h, k, \pm l$ were measured in the range $2\theta < 125^\circ$. The intensities of three standard reflections were measured after every 57 reflections; these intensities dropped by an average of a few percent over the period of data collection, but no correction factor was applied because the decrease was not considered significant. No absorption corrections were applied ($\mu = 6.24 \text{ cm}^{-1}$). Lorentz and polarization corrections were applied to convert intensities to structure amplitudes. Standard deviations in the intensities, $\sigma(I)$, and in the structure amplitudes, $\sigma(F_o)$, were derived directly from counting statistics.

Solution and Refinement of the Structure.

The structure was solved by direct methods using MULTAN [33] to calculate phases for $300|E|$ values greater than 1.52. The E map computed from the phase set with the largest combined figure of merit (2.85) revealed all non-hydrogen atoms except for one carbon of the phenyl ring. The subsequent difference Fourier map readily gave the remaining carbon atom. The 4187 reflections of the independent reflections for which $|F_o| > 3\sigma(F_o)$ were used by the full-matrix least-squares refinement of the structure. The function minimized was $\Sigma w(|F_o| - |F_c|)^2$, where w is the weight calculated as $1/\sigma^2(F_o)$. An initial refinement using individual isotropic temperature factors for the non-hydrogen atoms led to a conventional $R = \Sigma ||F_o| - |F_c|| / \Sigma |F_o|$ of 0.180. After the successive refinement with the anisotropic temperature factors ($R = 0.102$), all of the hydrogen atoms were readily located from the difference Fourier map. Included in the further refinement were anisotropic thermal parameters for the non-hydrogen atoms and isotropic thermal parameters for hydrogen atoms, which started with those of the carbon atoms to which they are attached. The refinement, after 4 cycles full-matrix least-squares calculation, had converged to a conventional R of 0.085 and a weighted $R_w = (\Sigma w(|F_o| - |F_c|)^2 / \Sigma w|F_o|^2)^{1/2}$ of 0.092 with a QOF of 3.50 (Quality of fit = $[\Sigma w(|F_o| - |F_c|)^2 / (N_{\text{obsd}} - N_{\text{para}})]^{1/2}$). The final positional and thermal parameters are given in Table 2. The atomic scattering factors for O, N, and C atoms were those of "International Tables for X-Ray Crystallography" [34]; for H, the scattering factors of Stewart, Davidson, and Simpson were used. Mathematical and computational details are noted elsewhere [36].

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