Synthesis and Structure of 2,3,3a,4,9,9a-Hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline and 5,7-Dibromo-2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline

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A novel fused oxonorbornyl ring system synthesized by reaction of substituted aniline and methacrolein dimer in acidic media has been characterized by single crystal X-ray diffraction. Two such structures are reported herein. These compounds represent the first reported structures of fused oxonorbornyl derivatives.

The title compounds were first synthesized in 1941 at Battelle Institute but remained nearly fifty years structurally uncharacterized until the present study. Addition of methacrolein dimer [1] to a solution of aniline in aqueous alcoholic hydrogen chloride leads to rapid formation of crystalline 2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline (I) (Figure 1). This compound was purified by repeated recrystallization from ethanol to form large colorless prismatic crystals. Bromination of I in carbon tetrachloride gives 5,7-dibromo-2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline (II) which crystallizes from ethanol as large well formed crystals having light brown transparent color. Unambiguous structural elucidation has only recently been accomplished and is reported herein. Because of inherent ambiguities in the nmr and ir spectra, X-ray structure determination was performed. Suitable single crystals of I and II were chosen, and were attached to the end of a glass fiber using fast drying epoxy glue. All analyses were then performed at room temperature using an Enraf-Nonius automated diffractometer with graphite-monochromatized MoKα radiation (λ = 0.71073 Å). Cell constants were determined using 25 reflections in a range of θ: (I) 4.08 ≤ θ ≤ 14.37° (II) 7.24 ≤ θ ≤ 12.33°. Intensity data were collected using Ω–2θ scans over the range 2 ≤ θ ≤ 25°. For I, 1129 independent reflections were collected over the range of 0 ≤ h ≤ 15, 0 ≤ k ≤ 18, 0 ≤ 1 ≤ 6; for II 2864 reflections having 0 ≤ h ≤ 12, 0 ≤ k ≤ 16, –23 ≤ 1 ≤ 23 were measured. Profile analysis was performed on all reflections [2,3]. Both sets of data were corrected for secondary extinction and for absorption [4] ranging from 0.91 to 1.00 for compound I and from 0.81 to 1.00 for II as well as for Lorentz and polarization effects. Both structures were solved by direct methods. Positions of bromine atoms for II were found by this manner and confirmed by solution of the Patterson function. Full-matrix least squares refinements and difference electron density map analyses were used to resolve the remaining atoms in the structure. All non-hydrogen atoms for both structures were refined anisotropically.

The H atoms were either located by analysis of difference electron density maps or had reasonable coordinates calculated and were not refined. Atomic scattering factors were taken from International Tables for X-ray Crystallography [5]. All calculations were performed using PDP-11 minicomputer and Enraf-Nonius (1983) SDP-Plus software [6].
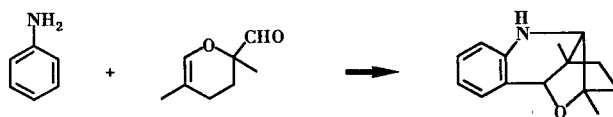

Figure 1. Formation of compound I.

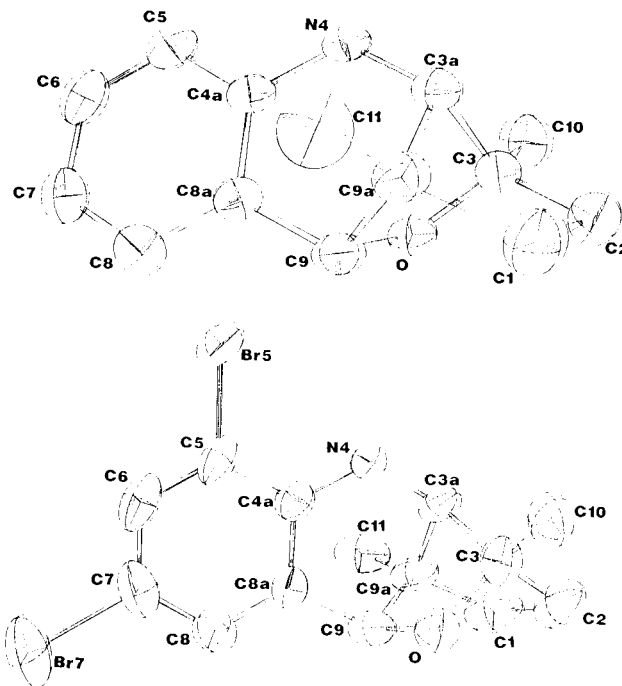

Figure 2. ORTEP representations with 50% probability ellipsoids and atom numbering schemes: a) I, 2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H- cyclopenta[b]quinoline; b) II, 5,7-dibromo-2,3,3a,4,9, 9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline.

I. Final refinement for 144 parameters yields R = 0.034, wR = 0.042; w = $1/\sigma^2(F_o)$, S = 1.378; (Δ/σ) max < 0.001. The largest peak in the resulting difference Fourier map was 0.116 eÅ^{-3}, and the deepest hole was –0.136 eÅ^{-3}. Final coordinates and their equivalent displacement parameters are given in Table 1; bond lengths and angles are supplied in Table 3. The molecule is illustrated and the labeling scheme is explained by the ORTEP representation [7] as shown in Figure 2(a).

Table 1

Positional Parameters (e.s.d.'s) and Equivalent Isotropic Thermal Parameters (Beq) for **I**

Atom	x	y	z	Beq(Å^2)
O	0.4685(1)	0.2694(1)	-0.020	3.12(3)
N4	0.4605(2)	0.2670(1)	0.4685(4)	3.15(4)
C1	0.6748(2)	0.2807(2)	0.0501(5)	4.16(6)
C2	0.6135(2)	0.3633(2)	-0.0052(5)	4.02(5)
C3	0.5115(2)	0.3440(1)	0.1064(4)	3.15(5)
C3a	0.5437(2)	0.2996(2)	0.3304(4)	3.03(5)
C4a	0.4181(2)	0.1852(2)	0.4215(4)	2.70(4)
C5	0.3501(2)	0.1455(2)	0.5716(5)	3.41(5)
C6	0.3083(2)	0.0643(2)	0.5168(5)	3.87(5)
C7	0.3329(2)	0.0217(2)	0.3161(5)	3.82(5)
C8	0.4015(2)	0.0606(2)	0.1685(5)	3.45(5)
C8a	0.4434(2)	0.1422(1)	0.2174(5)	2.72(4)
C9	0.5174(2)	0.1896(2)	0.0685(4)	2.96(5)
C9a	0.6055(2)	0.2267(2)	0.2109(5)	3.08(5)
C10	0.4337(2)	0.4163(2)	0.1128(6)	4.65(6)
C11	0.6586(2)	0.1610(2)	0.3614(6)	4.29(6)
H4	0.457	0.282	0.615	

II. The final refinement parameters for 325 parameters (two molecules per asymmetric unit) are R = 0.051, wR = 0.056; w = $1/\sigma^2(F_o)$, S = 1.342; (Δ/σ) max < 0.01. The largest remaining electron peaks were in the immediate vicinity of bromine atoms. The largest peak in the resulting difference Fourier map was 0.725 eÅ^{-3}, and the deepest hole was –0.536 eÅ^{-3}. Final coordinates and their equivalent displacement parameters for this structure are shown in Table 2. Bond lengths and angles are supplied in Table 3. An ORTEP representation [7] of the molecule is supplied in Figure 2(b) along with the atom labeling scheme.

Nearly all bond lengths and angles for both structures are in reasonable agreement with accepted values. The angle C9a–C3a–C3 at the bridgehead position is significantly smaller than anticipated, 94.1(2)° for **I** and 93.4(7), 95.2(7)° for **II**, but is consistent with the average angle of previously published structures that include a similar atomic arrangement [8-17]. Of interest is the significant difference in N4–C4a distance between the two structures 1.385(3) Å for **I**, and 1.358(11), 1.363(11) Å for **II**. For both compounds, the core geometrical arrangement of

Table 2

Positional Parameters (e.s.d.'s) and Equivalent Isotropic Thermal Parameters (Beq) for **II**

Atom	x	y	z	Beq(Å^2)
BR5	-0.0015(1)	0.2038(1)	0.15535(5)	6.60(3)
BR7	-0.3869(1)	0.41987(9)	0.25396(7)	6.88(3)
O	0.1360(6)	0.3211(4)	0.4251(3)	4.1(2)
N4	0.1036(7)	0.1888(5)	0.3128(3)	3.7(2)
C1	0.146(1)	0.1545(7)	0.5038(5)	5.8(3)
C2	0.268(1)	0.2100(7)	0.4962(5)	5.7(3)
C3	0.2373(9)	0.2502(7)	0.4229(5)	4.3(2)
C3a	0.1607(9)	0.1681(6)	0.3835(4)	3.5(2)
C4a	-0.0059(8)	0.2408(6)	0.2977(4)	3.3(2)
C5	-0.0705(8)	0.2569(7)	0.2301(5)	4.2(2)
C6	-0.1814(9)	0.3108(7)	0.2163(5)	4.7(2)
C7	-0.2321(9)	0.3483(7)	0.2715(5)	4.5(3)
C8	-0.1711(8)	0.3333(6)	0.3390(5)	3.8(2)
C8a	-0.0604(8)	0.2824(6)	0.3521(4)	3.1(2)
C9	0.0151(9)	0.2684(6)	0.4225(4)	3.7(2)
C9a	0.063(1)	0.1652(6)	0.4333(4)	4.1(2)
C10	0.3420(9)	0.2934(7)	0.3919(5)	4.9(3)
C11	-0.044(1)	0.0910(8)	0.4186(5)	5.9(3)
H4	0.138	0.162	0.275	
BR5'	0.6735(1)	0.48905(8)	0.47239(5)	5.19(3)
BR7'	0.6480(1)	0.09096(8)	0.50955(7)	6.41(3)
O'	0.8269(6)	0.3688(4)	0.7439(3)	4.0(1)
N4'	0.8399(7)	0.4763(5)	0.6206(4)	3.8(2)
C1'	1.083(1)	0.4158(8)	0.7762(5)	6.1(3)
C2'	0.983(1)	0.4702(8)	0.8090(5)	5.9(3)
C3'	0.8655(9)	0.4691(7)	0.7514(5)	4.1(2)
C3a'	0.9285(9)	0.4822(6)	0.6859(4)	3.8(2)
C4a'	0.8001(8)	0.3902(6)	0.5928(4)	3.2(2)
C5'	0.7228(8)	0.3788(7)	0.5285(4)	3.7(2)
C6'	0.6795(8)	0.2905(7)	0.5034(5)	4.4(2)
C7'	0.7136(9)	0.2113(7)	0.5421(5)	4.2(2)
C8'	0.7937(9)	0.2188(6)	0.6046(5)	4.0(2)
C8a'	0.8335(8)	0.3066(7)	0.6312(4)	3.6(2)
C9'	0.9092(9)	0.3225(6)	0.7023(5)	3.7(2)
C9a'	1.0158(9)	0.3951(7)	0.6996(5)	4.4(2)
C10'	0.753(1)	0.5300(7)	0.7602(5)	5.5(3)
C11'	1.106(1)	0.3763(8)	0.6475(6)	5.9(3)
H4'	0.807	0.533	0.597	

atoms is very similar. There are several planar moieties in the molecule, these are defined by C4a,C5,C6,C7,C8,C8a (plane 1), C4a,N4,C9,C8a (plane 2), C9a,C9,O,C3 (plane 3), C9a,C1,C2,C3 (plane 4) and C11,C9a,C3a,C3,C10 (plane 5). Formulae describing least-square planes and their angles of intersection are supplied in Table 4. Confirmation of existing strain in the molecule [18] is the bridgehead angle together with the flap angle between planes 3 and 4 similar to the flap angle in norbornane structures measuring from 108(2)° to 113(2)° [10-12,17]. To the best of authors' knowledge, the title compounds represent the first reported structures of fused oxonorbornyl derivatives. The most similar compounds that were found are longifolene [22] and 1,4-methano-5H-oxazolo[3,2-a]quinolin-2(1H)-one regioisomer [22]. Both reported structures

Table 3
Bond Distances (Å) and Angles (°) with e.s.d.'s for I and II

I / **II**

At. 1	At. 2	Distance	At. 1	At. 2	Distance	At. 1	At. 2	Distance
O	C3	1.462(3)	BR5	C5	1.883(10)	BR5'	C5'	1.917(9)
O	C9	1.462(3)	BR7	C7	1.901(9)	BR7'	C7'	1.895(9)
N4	C3a	1.449(3)	O	C3	1.466(11)	O'	C3'	1.466(11)
N4	C4a	1.385(3)	O	C9	1.469(11)	O'	C9'	1.436(11)
C1	C2	1.523(4)	N4	C3a	1.436(10)	N4'	C3a'	1.450(10)
C1	C9a	1.545(4)	N4	C4a	1.358(11)	N4'	C4a'	1.363(11)
C2	C3	1.524(4)	C1	C2	1.53(2)	C1'	C2'	1.53(2)
C3	C3a	1.532(4)	C1	C9a	1.509(12)	C1'	C9a'	1.568(13)
C3	C10	1.500(3)	C2	C3	1.517(13)	C2'	C3'	1.526(13)
C3a	C9a	1.539(3)	C3	C3a	1.539(13)	C3'	C3a'	1.541(13)
C4a	C5	1.391(3)	C3	C10	1.474(14)	C3'	C10'	1.492(15)
C4a	C8a	1.401(3)	C3a	C9a	1.529(14)	C3a'	C9a'	1.529(13)
C5	C6	1.384(3)	C4a	C5	1.397(11)	C4a'	C5'	1.384(11)
C6	C7	1.378(4)	C4a	C8a	1.411(12)	C4a'	C8a'	1.405(12)
C7	C8	1.383(4)	C5	C6	1.383(13)	C5'	C6'	1.383(13)
C8	C8a	1.382(3)	C6	C7	1.381(14)	C6'	C7'	1.358(13)
C8a	C9	1.491(3)	C7	C8	1.378(13)	C7'	C8'	1.367(12)
C9	C9a	1.537(3)	C8	C8a	1.358(12)	C8'	C8a'	1.376(13)
C9a	C11	1.501(4)	C8a	C9	1.480(11)	C8a'	C9'	1.495(11)
N4	H4	0.885	C9	C9a	1.538(13)	C9'	C9a'	1.526(13)
O	H4	2.15	C9a	C11	1.528(14)	C9a'	C11'	1.53(2)
			N4	H4	0.944	N4'	H4'	0.959
			O	H4'	2.19			

I / **II**

A. 1	A. 2	A. 3	Angle	A. 1	A. 2	A. 3	Angle	A. 1	A. 2	A. 3	Angle
C3	O	C9	106.8(1)	C3	O	C9	106.9(6)	C3'	O'	C9'	107.9(7)
C3a	N4	C4a	120.2(2)	C3a	N4	C4a	121.5(7)	C3a'	N4'	C4a'	120.7(7)
C2	C1	C9a	104.4(2)	C2	C1	C9a	103.6(8)	C2'	C1'	C9a'	104.7(8)
C1	C2	C3	102.8(2)	C1	C2	C3	103.1(8)	C1'	C2'	C3'	102.9(8)
O	C3	C2	105.9(2)	O	C3	C2	105.7(8)	O'	C3'	C2'	105.1(7)
O	C3	C3a	101.6(2)	O	C3	C3a	101.4(7)	O'	C3'	C3a	100.5(7)
O	C3	C10	108.0(2)	O	C3	C10	109.4(8)	O'	C3'	C10'	110.5(8)
C2	C3	C3a	101.9(2)	C2	C3	C3a	101.7(8)	C2'	C3'	C3a	101.4(8)
C2	C3	C10	118.4(2)	C2	C3	C10	118.6(8)	C2'	C3'	C10'	118.2(8)
C3a	C3	C10	119.2(2)	C3a	C3	C10	118.2(8)	C3a'	C3'	C10'	118.9(8)
N4	C3a	C3	114.6(2)	N4	C3a	C3	115.4(7)	N4'	C3a'	C3'	114.4(7)
N4	C3a	C9a	114.3(2)	N4	C3a	C9a	113.1(7)	N4'	C3a'	C9a'	113.0(7)
N3	C3a	C9a	94.1(2)	N3	C3a	C9a	93.4(7)	N3'	C3a'	C9a'	95.2(7)
N4	C4a	C5	121.4(2)	N4	C4a	C5	123.8(8)	N4'	C4a'	C5'	124.0(8)
N4	C4a	C8a	119.2(2)	N4	C4a	C8a	119.9(7)	N4'	C4a'	C8a'	119.8(7)
C5	C4a	C8a	119.5(2)	C5	C4a	C8a	116.2(8)	C5'	C4a'	C8a'	116.2(8)
C4a	C5	C6	119.6(2)	BR5	C5	C4a	118.1(7)	BR5'	C5'	C4a'	119.0(7)
C5	C6	C7	121.2(2)	BR5	C5	C6	119.3(7)	BR5'	C5'	C6'	118.7(6)
C7	C8	C8a	120.8(2)	C4a	C5	C6	122.6(9)	C4a'	C5'	C6'	122.3(8)
C4a	C8a	C8	119.7(2)	C5	C6	C7	118.8(8)	C5'	C6'	C7'	119.8(8)
C4a	C8a	C9	115.5(2)	BR7	C7	C6	119.6(7)	BR7'	C7'	C6'	119.8(7)
C8	C8a	C9	124.7(2)	BR7	C7	C8	120.3(8)	BR7'	C7'	C8'	120.3(7)
O	C9	C8a	108.3(2)	C6	C7	C8	120.0(8)	C6'	C7'	C8'	120.0(9)
O	C9	C9a	102.9(2)	C7	C8	C8a	120.8(9)	C7'	C8'	C8a'	120.6(8)
C8a	C9	C9a	110.7(2)	C8a	C9a	C8	121.4(7)	C8a'	C9a'	C8'	121.0(7)
C1	C9a	C3a	102.3(2)	C4a	C8a	C9	114.5(7)	C4a'	C8a'	C9'	114.6(8)
C1	C9a	C9	108.1(2)	C8	C8a	C9	124.1(8)	C8	C8a	C9	124.4(8)
C1	C9a	C11	115.5(2)	O	C9	C8a	107.3(7)	O'	C9'	C8a'	107.9(7)
C3a	C9a	C9	96.1(2)	O	C9	C9a	102.0(7)	O'	C9'	C9a'	103.6(7)
C3a	C9a	C11	117.1(2)	C8a	C9	C9a	111.4(7)	C8a'	C9'	C9a'	110.7(8)
C9	C9a	C11	115.4(2)	C1	C9a	C3a	103.0(8)	C1'	C9a'	C3a'	100.5(7)
N4	H4	O	158.9	C1	C9a	C9	110.2(7)	C1'	C9a'	C9'	108.2(8)

Table 3 (continued)

I

A. 1	A. 2	A. 3	Angle
C3a	N4	H4	119.8
C4a	N4	H4	113.2
C3	O	H4	117.5
C9	O	H4	117.0

II

A. 1	A. 2	A. 3	Angle	A. 1	A. 2	A. 3	Angle
C1	C9a	C11	113.9(8)	C1'	C9a'	C11'	115.3(8)
C3a	C9a	C9	97.4(7)	C3a'	C9a'	C9'	96.5(7)
C3a	C9a	C11	117.1(8)	C3a'	C9a'	C11'	116.7(8)
C9	C9a	C11	113.7(8)	C9'	C9a'	C11'	117.0(8)
C3	O	H4'	121.5	N4'	H4'	O	147.0
C9	O	H4'	130.1	C3a'	N4'	H4'	120.1
				C4a'	N4'	H4'	119.1

evidence intermolecular hydrogen bonding in the solid [21] between a hydrogen atom H4 on the nitrogen atom N4 and an oxygen atom O (or O' for **II**) from a neighboring molecule. The distance of this interaction is 2.15 Å for **I** and 2.19 Å for **II**.

Table 4

Angles Between Planes for compounds **I** and **II**

Plane No.	Plane No.	Dihedral Angle [°]		
		for **I**	for **II** [a]	
1	2	4.1	2.7	1.8
3	4	112.5	113.2	112.5
3	5	121.8	120.7	122.1
4	5	126.3	130.1	125.1
2	5	105.5	105.1	104.5

[a] There are two molecules per asymmetric unit for **II**. These are designated primed (') and unprimed. The angle between the two aromatic rings of **II** in the same asymmetric unit is 116.3°.

EXPERIMENTAL

Melting points were determined and corrected. The ir spectra were measured on a Mattson Galaxy 4020 FT-IR spectrophotometer as potassium bromide pellets. The nmr spectra were measured on 300 MHz Bruker spectrometer in deuteriochloroform solution. Both compounds gave satisfactory elemental analysis.

Compound **I**. 2,3,3a,4,9,9a-Hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline.

Slow addition of 163 g of aniline to 80 ml of concentrated hydrochloric acid followed by dilution with 160 ml of alcohol resulted in a clear yellow solution. At a temperature of 27° 136.5 g of methacrolein dimer (3,4-dihydro-2,5-dimethyl-1,2H-pyran-2-carboxaldehyde) was rapidly added dropwise, shaking and maintaining the temperature at 40-50° by frequently dipping the reaction flask in cold water. The product began to crystallize before all of the dimer was added, and the temperature rose to 63°. The mixture was cooled and the flask stoppered and allowed to stand overnight. The solid was separated from the viscous, green liquid by suction and pressing, washed with 150 ml of dilute (1:1) alcohol, and dried at room temperature, yielding 150.4 g (72% yield based on dimer), of small, light green crystals. This product was purified by repeated crystallization from ethanol (7 ml per g), mp 173-174°; ir: 3327 (NH), 3096, 3049 (aromatic CH), 2957,

2934, 2872 (aliphatic CH_2, CH_3), 1606 (aromatic C=C), 1490, 1459, 1374, 1328, 1281 (ether CO), 1158 (CN), 1112, 972, 927, 865, 803, 749 cm^{-1}; ^{1}H nmr: δ 0.93 (s, 3H, C11 H_3), 1.33 (s, 3H, C10 H_3), 1.64, 1.85 (m, 2 x 2H, C1 H_2, C2 H_2), 2.71 (s, 1H, C3a H), 4.20 (s, 1H, C9 H), 4.33 (broad s, 1H, N4 H), 6.48 (m, 2H, H6, H8), 6.82 (d, 1H, H5), 7.01 (t, 1H, H7); ^{13}C nmr: δ 15.39 (s, 1C, C11), 16.37 (s, 1C, C10), 32.00, 34.24 (2s, 2C, C1, C2), 37.31 (s, 1C, C9a), 64.40 (s, 1C, C3a), 84.88 (s, 1C, C9), 93.62 (s, 1C, C3), 111.60, 115.93 (2s, 2C, C6, C8), 125.17 (s, 1C, C8a), 126.22 (s, 1C, C5), 128.81 (s, 1C, C7), 143.30 (s, 1C, C4a).

Anal. Calcd. for $C_{14}H_{17}NO$: C, 78.10; H, 7.96; N, 6.51. Found: C, 78.38; H, 7.94; N, 6.4.

Compound **II**. 5,7-Dibromo-2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline.

Bromination of **1** in carbon tetrachloride or in presence of pyridine afforded compound **II**. It crystallizes from alcohol solution, mp 158-159°; ir: 3343 (NH), 3065 (aromatic CH), 2965, 2934, 2872 (aliphatic CH_2, CH_3), 1598 (aromatic C=C), 1490, 1382, 1289 (ether CO), 1166 (CN), 1119, 980, 934, 865, 811 cm^{-1}; ^{1}H nmr: δ 0.89 (s, 3H, C11 H_3), 1.32 (s, 3H, C10 H_3), 1.64, 1.86 (m, 2 x 2H, C1 H_2, C2 H_2), 2.89 (s, 1H, C3a H), 4.14 (s, 1H, C9 H), 4.75 (broad s, 1H, N4 H), 6.87 (d, 1H, H6), 7.35 (d, 1H, H8); ^{13}C nmr:

δ 15.15 (s, 1C, C11), 16.24 (s, 1C, C10), 31.67, 34.07 (2s, 2C, C1, C2), 37.48 (s, 1C, C9a), 64.63 (s, 1C, C3a), 83.96 (s, 1C, C9), 94.56 (s, 1C, C3), 106.22, 106.99 (2s, 2C, C6, C8), 127.76 (s, 1C, C8a), 128.09 (s, 1C, C5), 133.46 (s, 1C, C7), 139.89 (s, 1C, C4a).

Anal. Calcd. for $C_{14}H_{15}Br_2NO$: Br, 42.84. Found: Br, 42.70.

Crystal Data.

Compound **I**, 2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline, $C_{14}H_{17}NO$, Mr = 216.31, 0.34 x 0.39 x 0.42 mm, orthorhombic, Pna2$_1$, a = 13.196(6), b = 15.122(5), c = 5.850(1) Å, V = 1167(1) Å^3, Z = 4, Dm = 1.22 g cm^{-3}, Dx = 1.231 g cm^{-3}, λ(MoKα) = 0.71073 Å, μ = 0.781 cm^{-1}, F(000) = 464, T = 205K, R = 0.034, wR = 0.042 for 929 independent reflections with I $\geq 3\sigma$(I) and 144 variables.

Compound **II**, 5,7-dibromo-2,3,3a,4,9,9a-hexahydro-3,9a-epoxy-1H-cyclopenta[b]quinoline, $C_{14}H_{15}Br_2NO$, Mr = 373.087, 0.34 x 0.34 x 0.41 mm, monoclinic, P2$_1$/n, a = 10.557(2), b = 14.045(6), c = 19.440(8) Å, β = 99.47(2)°, V = 2843(3) Å^3, Z = 8, Dm = 1.74 g cm^{-3}, Dx = 1.743 g cm^{-3}, λ (MoKα) = 0.71073 Å, μ = 56.343 cm^{-1}, F(000) = 1472, T = 295K, R = 0.051, wR = 0.056 for 2208 independent reflections with I $\geq 2.5\sigma$(I) and 325 variables.

For each crystal, three standard reflections were monitored at one-hour intervals with no apparent degradation in crystallinity.

REFERENCES AND NOTES

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[1] G. G. Stoner and J. S. McNulty, *J. Am. Chem. Soc.*, **72**, 1531 (1950).

[2] D. F. Grant and E. J. Gabe, *J. Appl. Cryst.*, **11**, 114 (1978).

[3] M. S. Lehman and F. K. Larsen, *Acta Cryst.*, **A30**, 580 (1974).

[4] A. C. T. North, D. C. Phillips and F. S. Mathews, *Acta Cryst.*, **A24**, 351 (1968).

[5] D. T. Cromer and J. T. Waber, International Tables for X-ray Crystallography, Vol **IV**, Table 2.2B. Birmingham:Kynoch Press (Present Distributor Kulwer Academic Publishers, Dordrecht.), 1974.

[6] Enraf-Nonius, Structure Determination Package. Enraf-Nonius, Delft, The Netherlands, 1983.

[7] C. K. Johnson, ORTEP. Report ORNL-5138. Oak Ridge National Laboratory, Tennessee, USA, 1976.

[8] D. Allen, C. J. L. Lock and G. Turner, *Can. J. Chem.*, **53**, 2707 (1975).

[9] J. N. Brown, R. L. R. Towns and L. M. Trefonas, *J. Heterocyclic Chem.*, **7**, 1321 (1970).

[10] A. F. Cesur and D. F. Grant, *Acta Cryst.*, **18**, 55 (1965).

[11] J. F. Chiang, C. F. Wilcox Jr. and S. H. Bauer, *J. Am. Chem. Soc.*, **90**, 3149 (1968).

[12] J. P. Declercq, G. Germain and M. Meerssche, *Acta Cryst.*, **B31**, 2896 (1975).

[13] J. Estienne, G. Pepe, D. Siri, A. Baldy and A. Samat, *Acta Cryst.*, **C48**, 352 (1992).

[14] A. C. Macdonald and J. Troter, *Acta Cryst.*, **18**, 243 (1965).

[15] A. C. Macdonald and J. Troter, *Acta Cryst.*, **19**, 456 (1965).

[16] M. G. Newton, N. S. Pataleo, S. Kirbawy and N. L. Allinger, *J. Am. Chem. Soc.*, **100**, 2176 (1978).

[17] A. Yokozeki and K. Kuchitsu, *Bull. Chem. Soc. Japan*, **44**, 22356 (1971).

[18] C. Van Alsenoy, J. N. Scarsdale and L. Schafer, *J. Comput. Chem.*, **3**, 53 (1982).

[19] Y. Brunel, C. Coulombeau, C. Coulombeau, M. Moutin and H. Jobic, *J. Am. Chem. Soc.*, **105**, 6411 (1983).

[20] Y. Ishii, T. Noda, M. Kawahara and M. Ogawa, *Org. Magn. Reson.*, **22**, 416 (1984).

[21] W. C. Hamilton, Hydrogen Bonding in Solids, W. A. Benjamin, Inc., NY, 1968.

[22] A. F. Cesur and D. F. Grant, *Acta Cryst.*, **18**, 55 (1965).

[23] A. Padwa, H. L. Gingrich and R. Lim, *Tetrahedron Letters*, **21**, 3419 (1980).