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rel-(1*R*,4*S*,5*S*,8*R*)-1,2,3,4,5,6,7,8-Octahydro-1,4:5,8-dimethanonaphthalene-2,2,3,3-tetracarbonitrile, C₁₆H₁₂N₄

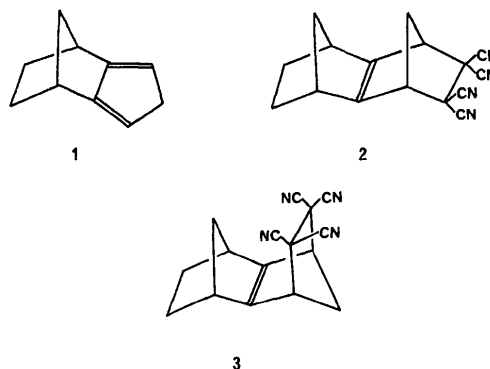BY WILLIAM H. WATSON, RAM P. KASHYAP,* PAUL D. BARTLETT AND CHENGJIU WU

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Abstract. $M_r = 260.30$, monoclinic, $P2_1/c$, $a = 18.123$ (5), $b = 13.509$ (3), $c = 12.073$ (2) Å, $\beta = 109.35$ (3)°, $V = 2789$ (1) Å³, $Z = 8$, $D_x = 1.240$ g cm⁻³, Cu $K\alpha$, $\lambda = 1.54178$ Å, $\mu = 6.24$ cm⁻¹, $F(000) = 1088$, room temperature, two independent molecules per asymmetric unit, $R = 0.043$ for 2497 unique reflections with $I > 3\sigma(I)$. The central π system in the *anti*-sesquinorbornene derivative deviates from planarity by approximately 4°. This is attributed primarily to an interaction between the central π bond and the π system of the cyano groups. The electron-withdrawing character of the cyano groups may account for the elongation of three C—C bonds in each molecule which range from 1.591 (3) to 1.609 (4) Å.

Introduction. Isodicyclopentadiene (1) reacts rapidly with tetracyanoethylene (TCNE) in benzene to yield a mixture of isomers (2) and (3) (Bartlett & Wu, 1984). When the reaction is followed by ¹H or ¹³C NMR it is found that (3) forms exclusively at the beginning, then slowly rearranges to (2). An 87:13 equilibrium mixture of 3:2 was obtained. To our knowledge, this is the first example in which a dienophile preferentially cycloadds to the *exo* face of (1) (Sugimoto, Kobuke & Furukawa, 1976; Paquette, Carr, Böhm & Gleiter, 1980; Watson, Galloy, Bartlett & Roof, 1981).

Experimental. Pyramidal-shaped crystal, $0.38 \times 0.30 \times 0.25$ mm, Syntex $P2_1$ diffractometer, $\theta:2\theta$ scan, variable scan rate, $2\theta_{max} = 120°$, graphite-monochromated Cu $K\alpha$ radiation; lattice parameters from

1

2

3

least squares of 15 medium-angle reflections with angles measured by centering routine associated with the diffractometer system (systematic absences $l = 2n + 1$ for $h0l$ and $k = 2n + 1$ for $0k0$); monitored reflection showed no change in intensity greater than $3\sigma(I)$; 3810 independent reflections ($0 \le h \le 19$, $0 \le k \le 14$, $-12 \le l \le 12$), 2497 with $I > 3\sigma(I)$; Lorentz and polarization, no absorption corrections; direct methods (*MULTAN78*: Main, Hull, Lessinger, Germain, Declercq & Woolfson, 1978) located all heavy atoms; hydrogen atom positions located in difference map but not refined in full-matrix least-squares anisotropic refinement; $\sum w(|F_o| - |F_c|)^2$ minimized, $w = 1/\sigma^2(F_o)$; 361 parameters refined; $R = 0.043$, $R_w = 0.048$, $S = 1.92$, $(\Delta/\sigma)_{av} = 0.057$, $(\Delta/\sigma)_{max} = 0.21$; highest peak in final difference map 0.15 e Å⁻³; locally written programs used for data reduction and initial block-diagonal least-squares refinement, *MULTAN78* for direct-methods calculations and *XRAY76* (Stewart,

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Table 1. *Fractional atomic coordinates* $(\times 10^4)$ *for non-hydrogen atoms and equivalent isotropic thermal parameters* $(\text{Å}^2 \times 10^3)$ *for the two independent molecules of compound* (3)

	x	y	z	U_{eq}*
Molecule 1				
C(1)	4636 (2)	4789 (2)	2258 (2)	46 (2)
C(2)	4091 (2)	4180 (2)	1164 (2)	44 (1)
C(3)	3906 (1)	3197 (2)	1771 (2)	42 (1)
C(4)	4355 (1)	3411 (2)	3132 (2)	45 (2)
C(4a)	5211 (1)	3387 (2)	3242 (2)	44 (1)
C(5)	5947 (2)	2777 (2)	3724 (3)	61 (2)
C(6)	6477 (2)	3393 (3)	4767 (3)	79 (2)
C(7)	6671 (2)	4327 (3)	4166 (3)	77 (2)
C(8)	6222 (2)	4156 (2)	2849 (2)	64 (2)
C(8a)	5368 (1)	4183 (2)	2735 (3)	46 (2)
C(9)	6318 (2)	3023 (3)	2775 (3)	75 (2)
C(10)	4224 (2)	4532 (2)	3150 (2)	49 (2)
C(11)	3390 (2)	4765 (2)	563 (3)	63 (2)
N(11)	2874 (2)	5259 (3)	120 (3)	105 (2)
C(12)	4478 (2)	3963 (2)	298 (3)	55 (2)
N(12)	4772 (2)	3810 (2)	−378 (3)	84 (2)
C(13)	3058 (2)	3084 (2)	1531 (2)	54 (2)
N(13)	2406 (2)	3012 (2)	1381 (2)	77 (2)
C(14)	4216 (2)	2303 (2)	1393 (2)	46 (2)
N(14)	4488 (2)	1617 (2)	1123 (2)	64 (2)
Molecule 2				
C(15)	771 (2)	3386 (2)	3914 (2)	49 (2)
C(16)	1235 (1)	3239 (2)	3012 (2)	45 (1)
C(17)	1031 (1)	4241 (2)	2250 (2)	44 (1)
C(18)	446 (2)	4784 (2)	2789 (3)	50 (2)
C(18a)	−264 (2)	4126 (2)	2513 (3)	49 (2)
C(19)	−1120 (2)	4077 (3)	1808 (3)	63 (2)
C(20)	−1556 (2)	4210 (3)	2712 (3)	71 (2)
C(21)	−1328 (2)	3275 (3)	3482 (3)	73 (2)
C(22)	−799 (2)	2689 (2)	2930 (3)	59 (2)
C(22a)	−79 (2)	3318 (2)	3173 (2)	46 (2)
C(23)	−1200 (2)	2954 (3)	1624 (3)	78 (2)
C(24)	852 (2)	4514 (2)	4086 (3)	60 (2)
C(25)	2082 (2)	3140 (3)	3645 (3)	62 (2)
N(25)	2721 (2)	3037 (3)	4167 (3)	100 (2)
C(26)	960 (2)	2349 (2)	2295 (3)	49 (2)
N(26)	715 (2)	1662 (2)	1749 (3)	72 (2)
C(27)	1719 (2)	4879 (2)	2423 (3)	54 (2)
N(27)	2223 (2)	5420 (2)	2593 (3)	81 (2)
C(28)	675 (2)	4045 (2)	987 (3)	50 (2)
N(28)	394 (2)	3913 (2)	9 (2)	74 (2)

* $U_{eq} = \frac{1}{3}[U_{11}(aa^*)^2 + U_{22}(bb^*)^2 + U_{33}(cc^*)^2 + 2U_{12}aba^*b^*\cos\gamma + 2U_{13}aca^*c^*\cos\beta + 2U_{23}bcb^*c^*\cos\alpha].$

Table 2. *Interatomic distances* (Å) *and valence angles* (°) *for compound* (3)

Molecule 1		Molecule 2	
C(1)—C(2)	1·591 (3)	C(15)—C(16)	1·594 (4)
C(1)—C(8a)	1·502 (4)	C(15)—C(22a)	1·506 (4)
C(1)—C(10)	1·540 (5)	C(15)—C(24)	1·538 (4)
C(2)—C(3)	1·605 (4)	C(16)—C(17)	1·609 (4)
C(2)—C(11)	1·469 (5)	C(16)—C(25)	1·477 (4)
C(2)—C(12)	1·468 (4)	C(16)—C(26)	1·469 (4)
C(3)—C(4)	1·600 (3)	C(17)—C(18)	1·595 (5)
C(3)—C(13)	1·474 (4)	C(17)—C(27)	1·473 (4)
C(3)—C(14)	1·467 (4)	C(17)—C(28)	1·470 (4)
C(4)—C(4a)	1·513 (3)	C(18)—C(18a)	1·508 (5)
C(4)—C(10)	1·534 (4)	C(18)—C(24)	1·537 (5)
C(4a)—C(5)	1·511 (4)	C(18a)—C(19)	1·503 (5)
C(4a)—C(8a)	1·314 (4)	C(18a)—C(22a)	1·327 (4)
C(5)—C(6)	1·549 (5)	C(19)—C(20)	1·555 (6)
C(5)—C(9)	1·545 (6)	C(19)—C(23)	1·533 (6)
C(6)—C(7)	1·553 (6)	C(20)—C(21)	1·541 (5)
C(7)—C(8)	1·545 (5)	C(21)—C(22)	1·554 (6)
C(8)—C(8a)	1·508 (4)	C(22)—C(22a)	1·502 (5)
C(8)—C(9)	1·546 (5)	C(22)—C(23)	1·544 (5)
C(11)—N(11)	1·129 (5)	C(25)—N(25)	1·129 (5)
C(12)—N(12)	1·132 (6)	C(26)—N(26)	1·140 (4)
C(13)—N(13)	1·139 (5)	C(27)—N(27)	1·134 (5)
C(14)—N(14)	1·147 (4)	C(28)—N(28)	1·134 (4)
C(2)C(1)C(8a)	105·2 (2)	C(16)C(15)C(22a)	104·7 (2)
C(2)C(1)C(10)	98·8 (2)	C(16)C(15)C(24)	99·6 (2)
C(8a)C(1)C(10)	100·4 (2)	C(22a)C(15)C(24)	100·0 (2)
C(1)C(2)C(3)	102·4 (2)	C(15)C(16)C(17)	102·3 (2)
C(1)C(2)C(11)	109·2 (2)	C(15)C(16)C(25)	110·6 (2)
C(1)C(2)C(12)	112·5 (3)	C(15)C(16)C(26)	110·6 (2)
C(3)C(2)C(11)	113·2 (3)	C(17)C(16)C(25)	112·0 (2)
C(3)C(2)C(12)	112·4 (2)	C(17)C(16)C(26)	112·5 (2)
C(11)C(2)C(12)	107·1 (2)	C(25)C(16)C(26)	108·8 (2)
C(2)C(3)C(4)	102·0 (2)	C(16)C(17)C(18)	102·2 (2)
C(2)C(3)C(13)	111·0 (2)	C(16)C(17)C(27)	112·8 (2)
C(2)C(3)C(14)	112·3 (2)	C(16)C(17)C(28)	112·4 (2)
C(4)C(3)C(13)	111·3 (2)	C(18)C(17)C(27)	108·6 (2)
C(4)C(3)C(14)	110·6 (2)	C(18)C(17)C(28)	112·6 (2)
C(13)C(3)C(14)	109·6 (2)	C(27)C(17)C(28)	108·2 (3)
C(3)C(4)C(4a)	104·1 (2)	C(17)C(18)C(18a)	105·8 (2)
C(3)C(4)C(10)	99·5 (2)	C(17)C(18)C(24)	98·5 (2)
C(4a)C(4)C(10)	100·3 (2)	C(18a)C(18)C(24)	99·8 (3)
C(4)C(4a)C(5)	142·6 (2)	C(18)C(18a)C(19)	142·8 (3)
C(4)C(4a)C(8a)	108·4 (2)	C(18)C(18a)C(22a)	109·0 (3)
C(5)C(4a)C(8a)	109·0 (2)	C(19)C(18a)C(22a)	108·2 (3)
C(4a)C(5)C(6)	104·5 (2)	C(18a)C(19)C(20)	105·5 (3)
C(4a)C(5)C(9)	98·2 (2)	C(18a)C(19)C(23)	99·0 (3)
C(6)C(5)C(9)	100·5 (3)	C(20)C(19)C(23)	99·8 (3)
C(5)C(6)C(7)	103·4 (3)	C(19)C(20)C(21)	103·4 (3)
C(6)C(7)C(8)	103·6 (3)	C(20)C(21)C(22)	103·8 (3)
C(7)C(8)C(8a)	105·4 (3)	C(21)C(22)C(22a)	104·3 (2)
C(7)C(8)C(9)	100·2 (3)	C(21)C(22)C(23)	99·9 (3)
C(8a)C(8)C(9)	98·6 (2)	C(22)C(22a)C(23)	98·8 (3)
C(1)C(8a)C(8)	142·9 (3)	C(15)C(22a)C(22)	143·3 (2)
C(1)C(8a)C(4a)	108·9 (2)	C(15)C(22a)C(18a)	108·2 (3)
C(4a)C(8a)C(8)	108·2 (2)	C(18a)C(22a)C(22)	108·5 (3)
C(5)C(9)C(8)	94·9 (3)	C(19)C(23)C(22)	95·3 (2)
C(1)C(10)C(4)	95·7 (2)	C(15)C(24)C(18)	96·1 (2)
C(2)C(11)N(11)	176·3 (3)	C(16)C(25)N(25)	176·7 (5)
C(2)C(12)N(12)	178·9 (8)	C(16)C(28)N(26)	176·8 (4)
C(3)C(13)N(13)	177·7 (4)	C(17)C(27)N(27)	174·9 (4)
C(3)C(14)N(14)	177·2 (4)	C(17)C(28)N(28)	178·6 (4)

Machin, Dickinson, Ammon, Heck & Flack, 1976) for all other computations; atomic scattering factors for C, O and N from Cromer & Mann (1968), for H from Stewart, Davidson & Simpson (1965). Table 1 lists atomic positional parameters and U_{eq} values while Table 2 gives bond lengths and valence angles.* The final structure suggests a possible crystallographic symmetry relationship between the two independent molecules; however, axial photographs, intensities of equivalent reflections and systematic extinctions are inconsistent with orthorhombic symmetry.

Discussion. Fig. 1 is an *ORTEP* (Johnson, 1971) drawing showing the two independent molecules of the

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* Lists of structure factors, hydrogen-atom coordinates and thermal parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 39150 (34 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.
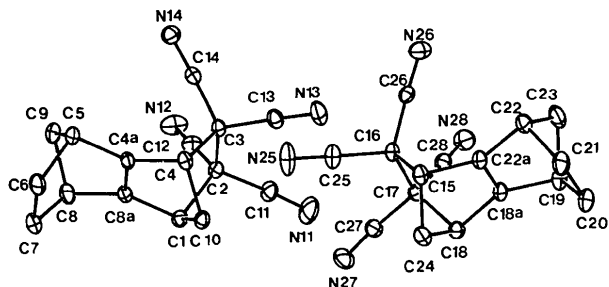

Fig. 1. *ORTEP* drawing of the two independent molecules of compound (3). Hydrogen atoms have been omitted for clarity. Thermal ellipsoids are drawn at the 35% probability level.

title compound (3). The molecules contain a non-crystallographic mirror plane bisecting the double bond and passing through the two methylene bridges. There is excellent agreement between equivalent bond distances related by the mirror plane and between the two independent molecules. There is little steric hindrance within the molecule and small conformational differences must be related to the packing environments of the two molecules. There are four intermolecular contacts between 2·25 (4) and 2·45 (4) Å involving hydrogen atoms.

Fig. 2 shows the closest intramolecular contacts and interplanar angles. The central π system in syn-sesquinorbornene derivatives is nonplanar and the interplanar angles equivalent to A,D in Fig. 2 range from 157·9 to 162·0° (Watson, Galloy, Bartlett & Roof, 1981; Pinkerton, Schwarzenbach, Stibbard, Carrupt & Vogel, 1981; Watson, Galloy, Combs & Bartlett, 1984). The deviations from planarity [pyramidalization of C(4a) and C(8a) types of carbon atoms] are attributed to ground-state torsional interactions between the π system and H(1), H(4), H(5) and H(8) (Houk, 1983; Houk, Rondan, Brown, Jorgensen, Mandura & Spellmeyer, 1983) or to repulsive hyperconjugative interactions between the π system and σ bonds (Gleiter & Böhm, 1983; Vogel, 1983; Spanget-Larsen & Gleiter, 1983). Hyperconjugative interactions have been attributed to interactions with the σ bonds of the ethylene bridge or to interactions with the methylene bridge. The assignments are related to the specific theoretical procedures used. The π system in anti-sesquinorbornenes is usually planar due to cancellation of torsional or hyperconjugative interactions in the two halves of the molecule.

The sesquinorbornene data are consistent with a low out-of-plane bending force constant about C(4a)—C(8a). In syn-sesquinorbornene the minimum does not coincide with the planar conformation due to asymmetric interactions about the double bond: however, these interactions are symmetric in most anti-sesquinorbornene derivatives. Although compound (3)
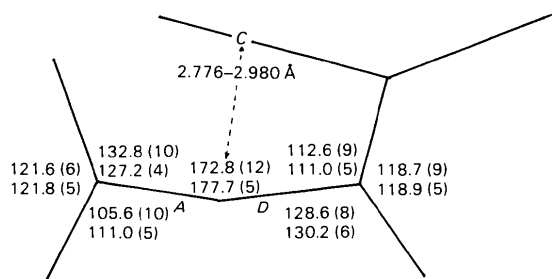
is an anti-sesquinorbornene derivative, the central double bond is nonplanar. The exact degree of bending is difficult to assess because planes A and D in molecule 1 show deviations from planarity of 0·06 (1) and 0·04 (1) Å. The central double-bond system probably deviates from planarity by about 5°. This deviation from planarity is attributed to the asymmetric environment of the double bond. Two rationalizations are possible. 1. The cyano groups increase the anti-hyperconjugative interactions of C(1)—C(2) and C(3)—C(4) [or possibly C(1)—C(10), C(12)—C(10)] with the π system. This may result from either better overlap because of the increase in the C(2)—C(3) distance or electronic modification of the C(1)—C(2) and C(3)—C(4) bonds. 2. The π system of two cyano groups and the π system of the central double bond interact repulsively. The C(4a)···C(14) and equivalent pair separations range from 2·777 (3) to 2·895 (6) Å. The interaction leads to a slight excess of π-electron density on the side opposite to the cyano groups. The latter effect is probably dominant.

The carbon atoms connected to the cyano groups are characterized by long C—C bonds. The C(1)—C(2), C(2)—C(3), C(3)—C(4) distances in molecules 1 and 2 are 1·591 (3), 1·605 (4), 1·600 (3) Å and 1·594 (4), 1·609 (4) and 1·595 (5) Å, respectively. This elongation was observed in a TCNE adduct of another isomer of isodicyclopentadiene (Kashyap, Watson, Wu & Bartlett, 1984) and is attributed to the electron-withdrawing character of the cyano groups. The C(sp^3)—C(sp^2) and C≡N distances check well with distances in reference compounds (Wang & Stucky, 1973; Karle, Flippen, Huisgen & Schug, 1975).

Fig. 2. Interplanar angles and separation distances for the C=C and C≡N π bonds for the two independent molecules of compound (3).

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rel-(1R,4S,4aR,7R)- (1) and rel-(1R,4S,4aS,7S)- (2) Isomers of 1,2,3,4,4a,5,6,7-Octahydro-1,4:4a,7-dimethanonaphthalene-5,5,6,6-tetracarbonitrile, $C_{16}H_{12}N_4$
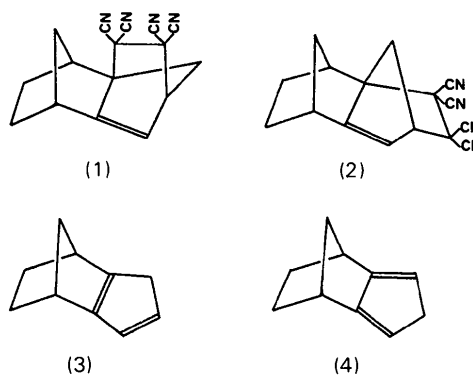
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Abstract. Compound (1): $M_r = 260.30$, monoclinic, $P2_1/c$, $a = 8.986$ (3), $b = 9.661$ (3), $c = 14.870$ (4) Å, $\beta = 94.86$ (2)°, $V = 1286.3$ (7) Å^3, $Z = 4$, $D_x = 1.344$ g cm^{-3}, Cu $K\alpha$, $\lambda = 1.54178$ Å, $\mu = 6.77$ cm^{-1}, $F(000) = 544$, room temperature, $R = 0.0397$ for 1494 unique reflections with $I > 3\sigma(I)$. Compound (2): $M_r = 260.30$, monoclinic, $P2_1/c$, $a = 9.696$ (2), $b = 8.925$ (1), $c = 16.022$ (4) Å, $\beta = 105.33$ (2)°, $V = 1337.1$ (5) Å^3, $Z = 4$, $D_x = 1.293$ Mg m^{-3}, Cu $K\alpha$, $\lambda = 1.54178$ Å, $\mu = 6.51$ cm^{-1}, $F(000) = 544$, room temperature, $R = 0.0345$ for 1447 unique reflections, $I > 3\sigma(I)$. The molecules are composed of end-to-edge fused norbornane systems. The sixteen $C(sp^3)$ valence angles of less than 105° imply considerable ring strain. C—C bond lengths of 1.577 (3) to 1.620 (3) Å are associated with the tetracyanoethylene moiety. The bond elongation is attributed to the electron-withdrawing character of the cyano groups.

(1)   (2)

(3)   (4)

Introduction. At elevated temperatures (3) is in equilibrium with isodicyclopentadiene (4) (Bartlett & Wu, 1983). Isomer (3) is more reactive and will preferentially form cycloadducts (Subramanyan, Bartlett, Iglesias, Watson & Galloy, 1982; Paquette, Williams, Carr, Charumilind & Blount, 1982). When isodicyclopentadiene (4) is added to a refluxing solution of tetracyanoethylene (TCNE) in m-dichlorobenzene, the

thermally stable products (1) and (2) are formed in a 3:1 ratio. Spectral data were consistent with a [6.2.1.1^{2,5}.0^{1,6}]dodec-6(7)-ene system which is confirmed by the present study. No cycloadducts of TCNE with (4) were found. Cycloadducts of TCNE with (4) are formed at lower temperatures; however, they undergo retro-Diels–Alder reactions even at room temperature. Adducts (1) and (2) can also be prepared by heating the adducts of (4) in m-dichlorobenzene at 423 K for 30 min (Bartlett & Wu, 1984).

Experimental. Syntex $P2_1$ diffractometer, $\theta:2\theta$ scan, variable scan rate, $2\theta_{max} = 120°$, graphite-monochromated Cu $K\alpha$ radiation; lattice parameters from least-squares refinement of 15 medium-angle reflections,

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