

Table 3. Comparison of the crystal data and the shortest intermolecular interactions (Å) of (TMDTDSF)₂NO₃ with those observed in (TMTTF)₂NO₃ and (TMTSF)₂NO₃

	(TMTTF) ₂ NO ₃ ^a TS = S	(TMDTDSF) ₂ NO ₃ ^b TS = S/Se	(TMTSF) ₂ NO ₃ ^c TS = Se
Crystal data			
<i>a</i> (Å)	7.061 (4)	7.155 (18)	7.127 (7)
<i>b</i> (Å)	7.395 (2)	7.472 (3)	7.567 (2)
<i>c</i> (Å)	12.547 (4)	12.661 (3)	12.822 (2)
α (°)	89.50 (3)	89.41 (3)	89.14 (2)
β (°)	93.91 (4)	86.17 (8)	86.61 (3)
γ (°)	108.97 (3)	70.90 (5)	70.56 (4)
<i>V</i> (Å ³)	618.2	638.1	659.1
Interplanar distances			
<i>d</i> 1	3.57	3.596 (7)	3.61
<i>d</i> 2	3.50	3.556 (7)	3.60
Intrastack contacts			
<i>d</i> 3: TS1—TS4 ⁱ	3.88 (2)	3.935 (2)	3.965
<i>d</i> 4: TS2—TS3 ⁱ	3.83 (2)	3.867 (2)	3.899
<i>d</i> 5: TS1—TS2 ⁱ	3.94 (2)	3.978 (2)	4.041
<i>d</i> 6: TS2—TS3 ⁱⁱ	3.78 (1)	3.853 (2)	3.913
<i>d</i> 7: TS1—TS4 ⁱⁱ	3.73 (1)	3.794 (2)	3.850
<i>d</i> 8: TS1—TS2 ⁱⁱ	4.00	4.057 (2)	4.120
Interstack contacts			
<i>d</i> 9: TS2—TS2 ⁱⁱⁱ	3.78	3.697 (2)	3.676
<i>d</i> 10: TS1—TS3 ^{iv}	3.94	3.832 (2)	3.822
<i>d</i> 11: TS3—TS3 ^{iv}	4.05	3.987 (2)	4.023

Symmetry code: (i) $1-x, 1-y, 1-z$; (ii) $-x, 1-y, 1-z$; (iii) $-x, 2-y, 1-z$; (iv) $1-x, -y, 1-z$.

References: (a) Liautard *et al.* (1982). (b) This work. (c) Soling *et al.* (1982); the e.s.d.'s of intermolecular Se...Se distances are 0.002–0.003 Å.

Maurin, 1982) and smaller than those of (TMTSF)₂NO₃ (Soling, Rindorf & Thorup, 1982). The bond distances, bond angles, interplanar separations, and intra- and interstack TS...TS contacts of (TMDTDSF)₂NO₃ are displayed in Figs. 1 and 2 and compared in Tables 2 and 3 to those observed in (TMTTF)₂NO₃ and (TMTSF)₂NO₃. From these comparisons, it appears that the TS—C bond distances, the C—TS—C bond angles and the inter-

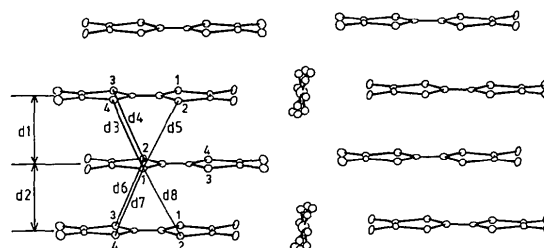


Fig. 2. Side view of stacks showing shorter S/Se...S/Se contacts corresponding to those given in Table 3.

molecular interactions are nearly midway between those observed in the TMTTF salt and those observed in the TMTSF salt. It is noted that the intrastack interactions increase while the interstack ones decrease from the TMTTF to the TMTSF salts.

The important disorder observed for the NO₃⁻ anion is the same as that described in detail in (TMTTF)₂NO₃ (Liautard *et al.*, 1982), *i.e.* the central N atom is surrounded by 12 statistically disordered O atoms in such a way that they form a planar disc. A dihedral angle of 103° is observed between the planar NO₃ and TMDTDSF counterparts.

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Acta Cryst. (1991). **C47**, 1943–1945

2,2-Dimethyl-1,2,3,4-tetrahydrobenzimidazo[3,2-a]pyrimid-4-one

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(Received 10 December 1990; accepted 1 March 1991)

Abstract. C₁₂H₁₃N₃O, *M*_r = 215.25, triclinic, *P* $\bar{1}$, *a* = 6.678 (3), *b* = 9.635 (3), *c* = 9.947 (5) Å, α = 114.79 (3), β = 106.01 (4), γ = 90.39 (4)°, *V* = 553.0 Å³, *Z* = 2, *D*_x = 1.29 Mg m⁻³, λ (Cu K α) = 1.5418 Å, μ = 0.655 mm⁻¹, *F*(000) = 228, *T* =

298 K, *R* = 0.036, *wR* = 0.036 for 1302 significant reflections out of a total 1383 [*I* > 2.5 σ (*I*)]. The molecular structure comprises a planar benzimidazole fused to a reduced pyrimidine ring, which contains an N atom bonded to an exocyclic H atom.

Introduction. One of us (Bird, 1965) reported the formation of the title compound by thermal rearrangement of 2-cyano-3,3-dimethyl-1-phenylpyrazolin-5-one. Structural confirmation by rational synthesis was not feasible. Thus an independent confirmation of the molecular structure was deemed desirable.

Experimental. A needle of dimensions 0.2 × 0.3 × 1.0 mm, prepared by the method of Bird (1965), crystallized from benzene was used on an automated Picker four-circle diffractometer using Ni-filtered Cu K α radiation and pulse-height analysis. Unit-cell dimensions were derived from 16 reflections $40 < 2\theta < 55^\circ$. The intensity of a standard reflection, measured every 25 reflections, showed no significant decay during data collection. Data were collected in the range $5 < 2\theta < 110^\circ$, each reflection being scanned over 2° in θ - 2θ scan mode. The index ranges were $h, -7$ to $6, k, 0$ to $10, l = -10$ to 9 . Friedel-related reflections were not measured. The E distribution was centric and phase sets in $P\bar{1}$ were generated by SOLVER (Gabe, Le Page, Charland, Lee & White, 1989). Only those phase sets with low figures of merit revealed a benzimidazole fragment, but none would refine. One fragment of the centrosymmetrically related pair was placed, with coordinates unchanged, in $P1$. This generated phases yielding a pair of fragments with a pseudo-centre at (0.529, 0.569, 0.580). The original fragment was moved (0.029, -0.069, -0.080) in $P\bar{1}$. This structure refined without problems. The positions of all H atoms were obtained from the ΔF map. Refinement was carried out on F^2 's using the NRCVAX (Gabe *et al.*, 1989) package, with non-H atoms refined anisotropically and H atoms isotropically; unit weights were used for refinement. The final R was 0.036, $wR = 0.036$, $w = 1/\sigma^2(F_o)$. The goodness of fit, S , was 6.9, the largest (shift/ σ) was 0.016 and the highest density on the ΔF map $0.14 \text{ e } \text{\AA}^{-3}$.

* Three very strong reflections, $12\bar{2}$ ($F_o = 108.0, F_c = 120.9$), 100 ($F_o = 45.5, F_c = 49.8$) and 110 ($F_o = 50.2, F_c = 57.9$) were excluded. This had no significant effect on any structural parameter.

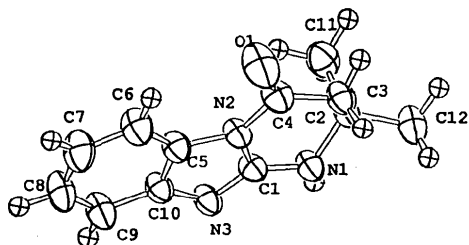


Fig. 1. ORTEP (Johnson, 1965) plot with 50% probability ellipsoids. For clarity, H atoms have been given arbitrary B isotropic thermal factors of 1.6 \AA^2 .

Table 1. Fractional atomic coordinates and equivalent isotropic temperature factors (\AA^2) with *e.s.d.*'s

$$B_{\text{eq}} = (8\pi^2/3) \sum_i U_{ij} a_i^* a_j^* a_i a_j$$

	x	y	z	B_{eq}
C1	0.7018 (3)	1.0478 (2)	0.4118 (2)	3.19 (10)
C2	0.7777 (3)	1.3056 (2)	0.6230 (2)	3.31 (10)
C3	0.5444 (4)	1.3159 (3)	0.5736 (3)	3.89 (12)
C4	0.4331 (3)	1.2212 (3)	0.4038 (3)	4.09 (12)
C5	0.4467 (3)	0.9680 (2)	0.1834 (2)	3.56 (11)
C6	0.2751 (4)	0.9451 (3)	0.0579 (3)	4.72 (13)
C7	0.2468 (4)	0.8088 (3)	-0.0754 (3)	5.27 (15)
C8	0.3860 (4)	0.7038 (3)	-0.0821 (3)	5.20 (14)
C9	0.5592 (4)	0.7284 (3)	0.0441 (3)	4.78 (13)
C10	0.5896 (3)	0.8634 (2)	0.1791 (2)	3.58 (11)
C11	0.8969 (4)	1.3838 (3)	0.5574 (3)	4.69 (15)
C12	0.8664 (5)	1.3790 (3)	0.7997 (3)	4.82 (15)
N1	0.8016 (3)	1.1404 (2)	0.5629 (2)	3.59 (9)
N2	0.5202 (2)	1.0891 (2)	0.3353 (2)	3.39 (9)
N3	0.7497 (3)	0.9161 (2)	0.3233 (2)	3.76 (9)
O1	0.2778 (3)	1.2510 (2)	0.3295 (2)	6.51 (10)
H1	0.926 (4)	1.121 (2)	0.597 (3)	4.3 (5)
H3A	0.520 (3)	1.418 (3)	0.601 (2)	3.9 (5)
H3B	0.469 (3)	1.281 (2)	0.630 (3)	4.5 (5)
H6	0.185 (4)	1.019 (3)	0.065 (3)	5.8 (6)
H7	0.130 (4)	0.786 (3)	-0.165 (3)	5.9 (6)
H8	0.366 (4)	0.612 (3)	-0.180 (3)	5.5 (6)
H9	0.661 (4)	0.662 (3)	0.036 (3)	5.7 (6)
H11A	1.056 (4)	1.369 (3)	0.581 (3)	7.0 (7)
H11B	0.887 (4)	1.498 (3)	0.596 (3)	7.0 (7)
H11C	0.840 (4)	1.333 (3)	0.433 (3)	6.6 (6)
H12A	0.791 (4)	1.329 (3)	0.842 (3)	7.1 (8)
H12B	1.013 (4)	1.373 (3)	0.828 (3)	5.7 (6)
H12C	0.847 (4)	1.493 (3)	0.845 (3)	6.3 (6)

Table 2. Bond lengths (\AA) and bond angles ($^\circ$)

C1—N1	1.347 (3)	C4—O1	1.210 (3)
C1—N2	1.400 (3)	C5—C6	1.377 (3)
C1—N3	1.308 (3)	C5—C10	1.388 (3)
C2—C3	1.516 (3)	C5—N2	1.411 (3)
C2—C11	1.522 (3)	C6—C7	1.385 (4)
C2—C12	1.522 (3)	C7—C8	1.372 (4)
C2—N1	1.474 (3)	C8—C9	1.384 (4)
C3—C4	1.497 (3)	C9—C10	1.385 (3)
C4—N2	1.387 (3)	C10—N3	1.412 (3)
N1—C1—N2	119.0 (2)	C6—C5—N2	132.0 (2)
N1—C1—N3	127.8 (2)	C10—C5—N2	105.1 (2)
N2—C1—N3	113.1 (2)	C5—C6—C7	116.7 (2)
C3—C2—C11	111.0 (2)	C6—C7—C8	121.3 (2)
C3—C2—C12	110.3 (2)	C7—C8—C9	121.7 (2)
C3—C2—N1	107.4 (2)	C8—C9—C10	118.1 (2)
C11—C2—C12	110.0 (2)	C1—N1—C2	118.1 (2)
C11—C2—N1	109.8 (2)	C5—C10—C9	119.3 (2)
C12—C2—N1	108.4 (2)	C5—C10—N3	111.0 (2)
C2—C3—C4	114.6 (2)	C9—C10—N3	129.7 (2)
C3—C4—N2	114.0 (2)	C1—N2—C4	124.6 (2)
C3—C4—O1	125.0 (2)	C1—N2—C5	106.0 (2)
N2—C4—O1	121.0 (2)	C4—N2—C5	129.3 (2)
C6—C5—C10	123.0 (2)	C1—N3—C10	104.8 (2)

Discussion. The molecular structure and atomic numbering scheme are shown in Fig. 1. Table 1* gives final fractional coordinates and equivalent isotropic thermal factors. Table 2 gives bond distances

* Lists of structure factors and anisotropic thermal parameters have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 54047 (11 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

and bond angles for non-H atoms. Bonds from carbon to hydrogen were in the range 0.93 (3) to 1.07 (3); N1 to H is 0.86 (2) Å. The molecular structure comprises a planar benzimidazole system fused to a reduced pyrimidine ring with two methyl substituents. The three atoms C1, N2 and N3 deviate by less than 0.05 Å from the best plane defined by the six benzene ring C atoms. Atom C1 is bonded to three N atoms. The bond lengths indicate a double bond to N3 [1.308 (3) Å] and single bonds to N1 [1.347 (3) Å] and N2 [1.400 (3) Å]. This bonding is in agreement with an H atom being bonded to N1 and not N3. Only atom C2 of the reduced pyrimidine ring is appreciably displaced from the plane occupied

by the remaining ring atoms. This is in marked contrast to the situation in the only other example of this ring system, 1,2,3,4-tetrahydro-1-methyl-2-oxo-pyrimido[1,2-*a*]benzimidazole-4-carboxylic acid methyl ester (Weber & Troxler, 1974).

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Acta Cryst. (1991). **C47**, 1945–1948

Structure du Chloro-3 Epoxy-4 β ,9 β Himachalanol-9a

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(Reçu le 22 février 1991, accepté 11 mars 1991)

Abstract. 3-Chloro-4 β ,9 β -epoxy-3,5,5,9-tetramethyl-decahydro-1*H*-benzocyclohepten-9a-ol, C₁₅H₂₅ClO₂, *M_r* = 272.82, monoclinic, *P*2₁, *a* = 17.811 (4), *b* = 10.734 (2), *c* = 18.061 (4) Å, β = 119.01 (2)°, *V* = 3019.7 Å³, *Z* = 8 (four molecules in the asymmetric unit), *D_x* = 1.201 g cm⁻³, λ (Cu *K* α) = 1.5418 Å, μ = 20.9 cm⁻¹, *F*(000) = 1232, *T* = 293 K, *R* = 0.050 for 9104 observed reflections. The structure has been elucidated and the absolute configuration determined. Three molecules exhibit the same conformation while the fourth shows some differences in the seven-membered ring. In the crystal, the molecules are linked in chains, through strong hydrogen bonds.

Introduction. La réactivité du β -himachalène (1), composant de l'huile essentielle du cèdre *Cedrus Atlantica*, et les propriétés de certains de ses dérivés sont décrites par ailleurs (Benharref, Chekroun & Lavergne, 1991). Si dans la plupart des cas, l'utilisation de la RMN 360 MHz à deux dimensions

a permis de déterminer la structure de ces composés, une analyse cristallographique a été nécessaire, pour identifier les produits de diépoxydation de ce sesquiterpène.

Comme le montre le schéma, l'action de l'acide métachloroperbenzoïque (*m*CPBA) sur (1), en quantité stoechiométrique, conduit avec un rendement quantitatif au seul α -6,7-époxyhimachal-2-ène, (2) (Bhan, Dev, Bass, Tagle & Clardy, 1982). En présence d'un excès de *m*CPBA, le β -himachalène donne un mélange de deux époxydes, (3) (30%) et (4) (70%). Un résultat identique est obtenu à partir de (2). Pour attribuer la stéréochimie du groupement époxy en position 2,3 dans les composés (3) et (4), le composé majoritaire (4) a été traité par HCl_{gaz} pour donner le 3-chloro 2,7-oxydohimachal-6-ol, (5). La détermination de la structure cristalline de ce composé a permis d'attribuer la stéréochimie des époxydes (3) et (4), et donc, celle de tous les dérivés préparés à partir de ces diépoxydes. Cette étude confirme la configuration absolue des himachalènes