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Structural Studies of Colchicine Derivatives. V. The Structure of 6,7-Didehydrocolchiceine, $C_{21}H_{21}NO_6$

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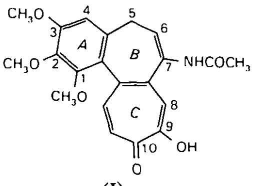
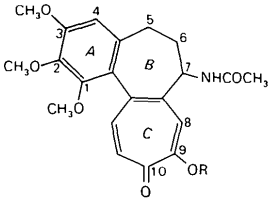
Abstract. $M_r = 383{\cdot}4$, monoclinic, $P2_1/a$, $a = 9{\cdot}535\,(6)$, $b = 16{\cdot}660\,(5)$, $c = 12{\cdot}450\,(4)$ Å, $\beta = 107{\cdot}89\,(6)°$, $V = 1882\,(1)$ Å^3, $Z = 4$, $D_x = 1{\cdot}353\,(1)$ g cm^{-3}, Mo $K\alpha$, $\lambda = 0{\cdot}71069$ Å, $\mu = 0{\cdot}93$ cm^{-1}, $F(000) = 808$, $T = 294$ K, final $R = 0{\cdot}040$ for 2016 unique observed reflections. The troponoid ring *C* presents an arrangement corresponding to an isocolchicine-like structure and shows a skew-boat conformation. The other seven-membered ring *B* is slightly flattened owing to the double bond and has a boat conformation. Three intramolecular hydrogen bonds influence the overall shape of the molecule.

Introduction. The title compound (I) is one of several colchiceine derivatives that has been synthetized and studied to find their conformation in order to correlate it with biological activity (Molins, Rius, Solans, Miravitlles, Bladé-Fonte & Germain, 1985, and references therein). Colchicine and some of its derivatives, which bind well to tubulin, behave as powerful antimitotics and, therefore, have potential antitumour activity.

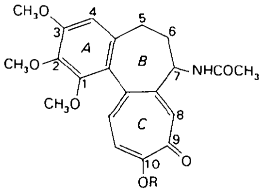
Isocolchicine and its derivatives (II) present considerably less activity, a fact that is strongly related to the



(I)



(II)



(III)

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arrangement of the tropolone ether function. In the present case we have undertaken the X-ray analysis of 6-dehydrocolchiceine to determine the arrangement of the oxygen functions of ring *C* and the general conformation of the molecule.

Experimental. Title compound obtained according to previous reported method (Bladé-Font, 1977). Single yellow crystals obtained by slow evaporation of ethyl acetate dilute solution. Size of crystal 0·2 × 0·2 × 0·3 mm. CAD-4 diffractometer. Unit-cell parameters by least squares from 25 reflections with θ between 4 and 12°. 3605 reflections collected, 3307 unique, 2016 considered observed with $I > 2\sigma(I)$; ω–2θ scan mode, *hkl* range: $-11 < h < 11$, $0 < k < 19$, $0 < l < 14$, θ limit 25°; scan range $\Delta\theta = (0{\cdot}80 + 0{\cdot}34\tan\theta)°$ extended 25% on either side of peak, max. scan time 60 s, three standard reflections recorded every hour and three orientation control reflections every 50 measurements. Variation of intensity in standard reflections 0·5%. Standard deviations determined from counting statistics. Lorentz–polarization corrections, not absorption. Structure solved by direct methods. Correct orientation of skeleton determined using *ROTSEARCH* (Rius & Miravitlles, 1984) and introduced in *MULTAN*11/82 (Main *et al.*, 1982). Anisotropic full-matrix least-squares refinement based on $|F_{hkl}|$ using *SHELX*76 (Sheldrick, 1976). All H-atom positions from $\Delta\rho$ synthesis, refined with fixed isotropic temperature parameters [methyl H atoms: $U = 0{\cdot}0979\,(33)$ Å^2, remaining H atoms: $U = 0{\cdot}0585\,(27)$ Å^2]. 2016 reflections included in final refinement cycle, 318 parameters refined; $R = 0{\cdot}040$, $wR = 0{\cdot}041$; $w = 2{\cdot}285/[\sigma^2(F) + 0{\cdot}00032F^2]$. Max. $(\Delta/\sigma) = 0{\cdot}26$. Scattering factors for non-hydrogen atoms from *International Tables for X-ray Crystallography* (1974). Residual electron density within $\pm 0{\cdot}18$ e Å^{-3}. Drawing by *PLUTO* (Motherwell & Clegg, 1978).

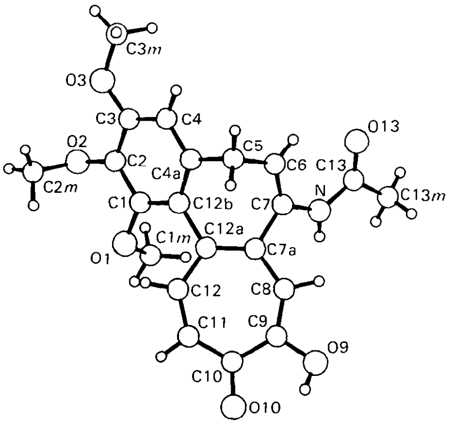

Fig. 1. A view of the molecule with the atomic numbering.

Table 1. *Atomic coordinates* ($\times 10^4$) (*with e.s.d.'s in parentheses*) *and equivalent isotropic temperature factors* (Å^2)

$$B_{\text{eq}} = (8/3)\pi^2 \textstyle\sum_i \sum_j U_{ij} a_i^* a_j^* \mathbf{a}_i.\mathbf{a}_j.$$

	x	*y*	*z*	B_{eq}
C(1)	−1999 (3)	−836 (1)	−4895 (2)	3·46
C(1*m*)	−3642 (4)	−467 (2)	−3903 (3)	5·94
C(2)	−2621 (3)	−1226 (2)	−5914 (2)	3·67
C(2*m*)	−3995 (4)	−390 (2)	−7382 (3)	5·15
C(3)	−1880 (3)	−1859 (2)	−6235 (2)	3·79
C(3*m*)	−1928 (4)	−2902 (2)	−7570 (3)	5·53
C(4)	−484 (3)	−2065 (2)	−5542 (2)	3·76
C(4a)	157 (3)	−1659 (2)	−4535 (2)	3·60
C(5)	1643 (3)	−1911 (2)	−3746 (3)	4·48
C(6)	1315 (3)	−2335 (2)	−2794 (2)	4·50
C(7)	786 (3)	−1945 (2)	−2071 (2)	3·81
C(7a)	666 (3)	−1052 (1)	−2063 (2)	3·50
C(8)	1315 (3)	−722 (2)	−966 (2)	3·79
C(9)	1440 (3)	33 (2)	−524 (2)	3·99
C(10)	780 (3)	773 (2)	−1076 (3)	4·17
C(11)	156 (3)	801 (2)	−2282 (2)	3·86
C(12)	−107 (3)	229 (2)	−3093 (2)	3·72
C(12a)	30 (3)	−630 (1)	−3061 (2)	3·45
C(12b)	−600 (3)	−1045 (1)	−4167 (2)	3·38
C(13)	514 (4)	−3096 (2)	−884 (2)	4·87
C(13*m*)	−19 (5)	−3317 (2)	80 (3)	6·18
O(1)	−2808 (2)	−223 (1)	−4628 (1)	4·10
O(2)	−4015 (2)	−1015 (1)	−6598 (2)	4·44
O(3)	−2605 (2)	−2216 (1)	−7237 (2)	4·86
O(9)	2222 (2)	98 (1)	579 (2)	5·31
O(10)	850 (3)	1384 (1)	−479 (2)	5·62
O(13)	1065 (3)	−3591 (1)	−1367 (2)	7·81
N	354 (3)	−2321 (1)	−1199 (2)	4·11

Table 2. *Bond lengths* (Å) *and bond angles* (°)

C(2)–C(1)	1·385 (4)	C(7a)–C(7)	1·493 (3)
C(12b)–C(1)	1·407 (3)	N–C(7)	1·420 (3)
O(1)–C(1)	1·381 (3)	C(8)–C(7a)	1·424 (4)
O(1)–C(1*m*)	1·433 (4)	C(12a)–C(7a)	1·394 (3)
C(3)–C(2)	1·395 (3)	C(9)–C(8)	1·363 (4)
O(2)–C(2)	1·386 (3)	C(10)–C(9)	1·457 (4)
O(2)–C(2*m*)	1·431 (4)	O(9)–C(9)	1·351 (3)
C(4)–C(3)	1·388 (4)	C(11)–C(10)	1·435 (4)
O(3)–C(3)	1·363 (3)	O(10)–C(10)	1·250 (3)
O(3)–C(3*m*)	1·435 (4)	C(12)–C(11)	1·355 (4)
C(4a)–C(4)	1·390 (4)	C(12a)–C(12)	1·435 (4)
C(5)–C(4a)	1·515 (4)	C(12b)–C(12a)	1·492 (3)
C(12b)–C(4a)	1·407 (3)	C(13*m*)–C(13)	1·486 (5)
C(6)–C(5)	1·494 (4)	O(13)–C(13)	1·229 (3)
C(7)–C(6)	1·329 (4)	N–C(13)	1·345 (3)

C(12b)–C(1)–C(2)	121·9 (2)	C(10)–C(9)–C(8)	128·2 (3)
O(1)–C(1)–C(2)	117·0 (2)	O(9)–C(9)–C(8)	116·0 (2)
O(1)–C(1)–C(12b)	121·1 (2)	O(9)–C(9)–C(10)	115·8 (2)
C(3)–C(2)–C(1)	120·3 (2)	C(11)–C(10)–C(9)	120·1 (2)
O(2)–C(2)–C(1)	120·3 (2)	O(10)–C(10)–C(9)	118·2 (3)
O(2)–C(2)–C(3)	119·4 (2)	O(10)–C(10)–C(11)	121·6 (3)
C(4)–C(3)–C(2)	118·9 (2)	C(12)–C(11)–C(10)	132·7 (3)
O(3)–C(3)–C(2)	115·6 (2)	C(12a)–C(12)–C(11)	133·3 (3)
O(3)–C(3)–C(4)	125·5 (2)	C(12)–C(12a)–C(7a)	122·5 (2)
C(4a)–C(4)–C(3)	120·8 (2)	C(12b)–C(12a)–C(7a)	122·0 (2)
C(5)–C(4a)–C(4)	120·9 (2)	C(12b)–C(12a)–C(12)	115·4 (2)
C(12b)–C(4a)–C(4)	121·4 (2)	C(4a)–C(12b)–C(1)	116·7 (2)
C(12b)–C(4a)–C(5)	117·5 (2)	C(12a)–C(12b)–C(1)	120·9 (2)
C(6)–C(5)–C(4a)	105·3 (2)	C(12a)–C(12b)–C(4a)	122·4 (2)
C(7)–C(6)–C(5)	121·4 (3)	O(13)–C(13)–C(13*m*)	122·2 (3)
C(7a)–C(7)–C(6)	122·6 (3)	N–C(13)–C(13*m*)	155·9 (3)
N–C(7)–C(6)	124·1 (2)	N–C(13)–O(13)	121·9 (3)
N–C(7)–C(7a)	113·2 (2)	C(2*m*)–O(2)–C(2)	113·1 (2)
C(8)–C(7a)–C(7)	112·3 (2)	C(3*m*)–O(3)–C(3)	117·7 (2)
C(12a)–C(7a)–C(7)	120·6 (2)	C(1*m*)–O(1)–C(1)	113·6 (2)
C(12a)–C(7a)–C(8)	127·0 (2)	C(13)–N–C(7)	127·8 (3)
C(9)–C(8)–C(7a)	134·6 (3)		

Discussion. Final atomic coordinates and equivalent isotropic thermal parameters are listed in Table 1;* Fig. 1 shows a view of the molecule with the atom numbering. The bond lengths and bond angles are listed in Table 2.

The ring *C* is not planar [r.m.s.d. = 0·05 (2) Å, $\delta_{\max}$ = 0·083 (7) Å for C(10)] and the bond lengths alternate corresponding to the isocolchicine form (II). Up to now the only colchiceine derivative that is totally in the colchicine form (III) in the solid state is surprisingly colchiceine itself (Silverton, 1979). The *C* ring shows a skew-boat conformation with C(10) at the prow of the boat as in colchiceine acetate (Miravitlles, Solans, Bladé-Font, Germain & Declercq, 1982), 7-deacetamidocolchiceine (Rius, Molins, Miravitlles & Bladé-Font, 1984) and colchiceine benzoate (Molins *et al.*, 1985).

The conformation of ring *B* differs from the colchicine derivatives, being definitely more flattened, as can be seen from the sum of the torsion angles of the ring (colchicine = 305°, isocolchicine = 300°, colchiceine acetate = 291°, colchiceine = 296°, 7-deacetamidocolchiceine = 292°, colchiceine benzoate = 298°), which have values around 300° in contrast to 6,7-didehydrocolchiceine (248°) and acetylanhydrocolchicine (248)°. In the latter, however, the flattening is caused by the additional pyrrolic ring, but in the case 6,7-didehydrocolchiceine, it is due to the C(6)—C(7) double bond. Therefore, comparison of the ring *B* torsion angles (Table 3) shows that the angle defined by C(7a)—C(7)—C(6)—C(5) is small in products (I) and (II) and, from the remaining torsion angles, the rest of the ring presents a conformation similar to those of the other colchicine derivatives. Thus, ring *B* has a boat conformation defined by the mean planes (1): C(4a), C(5), C(6); (2): C(4a), C(6), C(7), C(12b) [r.m.s.d.: 0·02 (2) Å]; and (3): C(7), C(7a), C(12a), C(12b) [r.m.s.d.: 0·007 (2) Å]. The dihedral angle between planes (1) and (2) is 56·3° and that between planes (2) and (3) is 39·6 (4)°.

Table 3. *Torsion angles in ring B* (°)

The maximum value of the e.s.d.'s is 2°.

	(I)	(II)	(III)		(IV)		(V)	(VI)	(VII)	(VIII)
C(4a)—C(12b)—C(12a)—C(7a)	−47	−53	−53	−57	−53	−53	−56	49	−52	−52
C(12b)—C(12a)—C(7a)—C(7)	2	24	−6	−7	−5	−5	4	3	−2	−2
C(12a)—C(7a)—C(7)—C(6)	49	19	78	79	79	81	75	−79	79	78
C(7a)—C(7)—C(6)—C(5)	−8	8	−44	−46	−48	−49	−54	49	−47	−49
C(7)—C(6)—C(5)—C(4a)	−68	−70	−44	−42	−43	−42	−36	39	−41	−39
C(6)—C(5)—C(4a)—C(12b)	70	72	68	70	73	70	69	−72	70	70
C(5)—C(4a)—C(12b)—C(12a)	2	3	7	9	4	5	2	1	3	1

(I) 6,7-Didehydrocolchiceine: present work; (II) acetylanhydrocolchicine (Miravitlles, Rius, Bladé-Font & Germain, 1983); (III) isocolchicine (Lessinger & Margulis, 1978*b*); (IV) colchicine (Lessinger & Margulis, 1978*a*); (V) colchiceine (Silverton, 1979); (VI) 7-deacetamidocolchiceine (Rius, Molins, Miravitlles & Bladé-Font, 1984); (VII) colchiceine benzoate (Molins *et al.*, 1985) and (VIII) colchiceine acetate (Miravitlles *et al.*, 1982).

The benzene ring *A* is planar {r.m.s.d.: 0·014 (6) Å, $\chi^2 = 6{\cdot}0$ [$=\sum(\Delta/\sigma)^2$], $\delta_{\max}$ = 0·018 (8) Å for C(4a)}. The methoxy group C(3)—O(3)—C(3*m*) is approximately in the plane of the benzene ring [5·0 (3)°], while the C(2)—O(2)—C(2*m*) group forms a dihedral angle of 91·6 (3)° with the ring. The C(1)—O(1)—C(1*m*) methoxy group, pointing in the opposite direction to C(2)—O(2)—C(2*m*), forms a dihedral angle of 84·3 (3)° with ring *A*. This conformation of the methoxy groups is similar to that of 7-deacetamidocolchiceine and acetylanhydrocolchicine. The *N*-acetyle group lies in a planar region [mean plane *D*: r.m.s.: 0·006 (3) Å, $\delta_{\max}$ = 0·010 (7) Å for N].

The general conformation of the molecule is defined by the dihedral angles between the planar regions and by the twisting about the C(12a)—C(12b) bond: ring *A*–ring *C*: 43·1 (3)°, ring *A*–group *D*: 74·3 (3)° and ring *C*–group *D*: 48·2 (3)°. The torsion angle C(12)—C(12a)—C(12b)—C(1) is −45·4 (4)°, being between −52 and −57° in all other colchicine derivatives studied so far.

The conformation is influenced by three intramolecular hydrogen bonds. The short O(1)···C(12) interaction may contribute to the low value of the C(12)—C(12a)—C(12b)—C(1) torsion angle. Similarly, the C(6)—C(7)—N—C(13) torsion angle is also small owing to the C(6)···O(13) intramolecular hydrogen bond. The H(9) atom appears to form a bifurcated hydrogen bond intramolecularly with O(10) and intermolecularly with O(13). An intermolecular bond has been observed between atoms O(10) and N. The packing of the molecules is stabilized through the hydrogen bonds.

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* Lists of anisotropic thermal parameters, structure factors, H-atom coordinates, torsion angles and hydrogen-bond dimensions have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 42185 (17 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

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Structures of the Monohydrated (I) and Anhydrous (II) Forms of 2,2′-[Benzylidenemethylenebis(carbonylamino)]dibenzoic Acid, $C_{24}H_{18}N_2O_6.H_2O$ and $C_{24}H_{18}N_2O_6$

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Abstract. (I) $M_r = 448.4$, triclinic, $P\bar{1}$, $a = 11.687$ (4), $b = 10.088$ (3), $c = 10.519$ (5) Å, $\alpha = 105.79$ (4), $\beta = 110.25$ (4), $\gamma = 97.83$ (3)°, $V = 1082.5$ Å^3, $Z = 2$, $D_x = 1.376$ Mg m^{-3}, λ(Cu $K\alpha$) = 1.54178 Å, $\mu = 0.82$ mm^{-1}, $F(000) = 468$, $T = 293$ K, $R(F) = 0.064$ for 2873 unique reflections with $F_o > 3\sigma(F_o)$. (II) $M_r = 430.4$, monoclinic, $P2_1/a$, $a = 16.469$ (3), $b = 11.334$ (6), $c = 11.312$ (3) Å, $\beta = 99.49$ (2)°, $V = 2082.6$ Å^3, $Z = 4$, $D_x = 1.373$ Mg m^{-3}, λ(Cu $K\alpha$) = 1.54178 Å, $\mu = 0.79$ mm^{-1}, $F(000) = 896$, $T = 293$ K, $R(F) = 0.071$ for 2726 unique reflections with $F_o > 3\sigma(F_o)$. The molecular conformations of (I) and (II) are different [the main difference involves rotation about one of the central C(=O)—C bonds: torsion angle −166.8 (3)° for (I) and −29.9 (5)° for (II)]. N—H···O intramolecular hydrogen bonds are formed between the NH and COOH groups in both molecules. One of the NH groups of (I) forms a bifurcated intramolecular hydrogen bond.

Introduction. The crystal structures of the title compounds (I) and (II) are of current interest because of the unusual melting behavior of crystal (I). The monohydrate crystal (I) obtained from methanol–water solution showed an apparent increase of the melting point from 468 to 513 K resulting from the preceding press or granulation processing applied to the single crystal. From the differential thermal analysis of the pressed monohydrate crystal, the recrystallization of the anhydrous crystallites seems to occur in the melting process followed by dehydration during the temperature increase. The anhydrous crystals (II), however, were obtained from acetic acid solution, and their melting point was observed as 513 K. In the present paper, we describe the crystal structures of (I) and (II), and discuss the differences found between them.

Experimental. For (I) and (II): D_m not determined; Rigaku AFC-5RU diffractometer, graphite-monochromated Cu $K\alpha$ radiation; 2θ–ω mode, $2\theta_{max} = 120°$; no significant intensity variation for three reflections monitored every 56 reflections; no absorption or extinction corrections; structures solved with *MULTAN78* (Main, Hull, Lessinger, Germain, Declercq & Woolfson, 1978); all H atoms located in difference Fourier maps; refinement by full-matrix least squares using modified version of *ORFLS* (Busing, Martin & Levy, 1962); non-H atoms anisotropic, H atoms isotropic; $\sum w(|F_o| - |F_c|)^2$ minimized with $w = 1/[\sigma^2(F) + (0.023F)^2]$; atomic scattering factors from *International Tables for X-ray Crystallography* (1974). (I): crystal 0.18 × 0.20 × 0.27 mm, 2873 reflections, $-13 \le h \le 12$, $0 \le k \le 11$, $-11 \le l \le 10$, $R = 0.064$, $wR = 0.060$, $(\Delta/\sigma)_{max} = 0.77$, no peaks greater than 0.18 e Å^{-3} in final difference Fourier map. (II): crystal 0.10 × 0.17 × 0.20 mm, 2726 reflections, $-18 \le h \le 16$, $0 \le k \le 12$, $0 \le l \le 12$, $R = 0.071$, $wR = 0.069$, $(\Delta/\sigma)_{max} = 0.9$, no peaks greater than