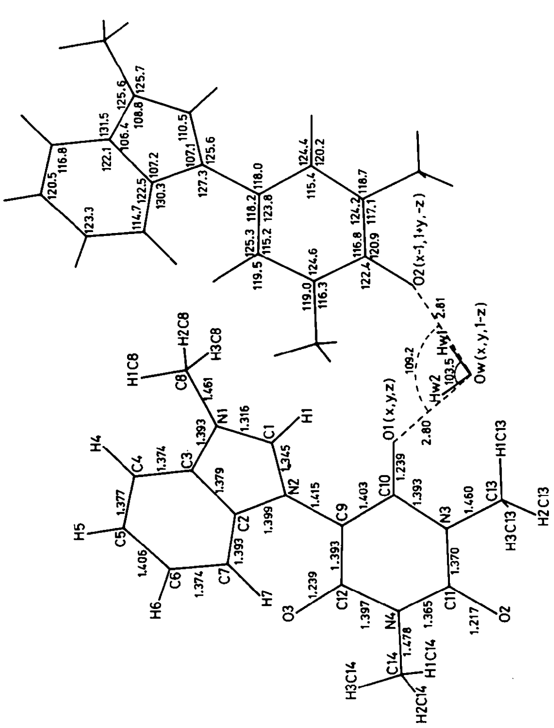


Fig. 1. Interatomic distances (Å) and angles (°) for the non-hydrogen atoms. Standard deviations are 0·004 Å in distances and 0·3° in angles.

The bond lengths and angles found in this structure are similar to those found in the structure of 5-(6-bromo-3-ethyl-2-methylbenzimidazolio)-4-barbiturate hydrate (Matthews, 1965). The N(2)—C(9) length of 1·415 (4) Å indicates considerable double-bond character and suggests significant conjugation between the benzimidazole and barbiturate ring systems. The bond lengths of the imidazole ring in the region of N(1) are similar to those found in a number of structures where the benzimidazole moiety is present (Quick & Williams, 1976, and references therein) but where it does not have a substituent bonded to N(1). This suggests that the majority of the positive charge is localized on N(1), and that the canonical form (I) is a principal contributor to the resonance hybrid so far as the benzimidazole moiety is concerned. The bond lengths C(12)—O(3) and C(10)—O(1) are equal and are significantly longer than C(11)—O(2) indicating that (I) and the canonical structure with the negative charge on O(3) contribute equally to the resonance hybrid so far as the barbiturate moiety is concerned. The benzene ring shows a large distortion from a regular hexagonal shape, being elongated along the C(4)—C(7) line in much the same manner as occurs in the bromo derivative referred to above. The benzimidazole ring and the barbiturate ring are both planar within experimental error, with the dihedral angle between the two planes being 121·6 (3)°.

We wish to thank Professor J. W. Clark-Lewis for supplying the crystals of the compound, Dr Y. Le Page of the National Research Council, Canada, for useful discussions, and Dr G. T. DeTitta for assistance in applying *QTAN* to this problem. One of the authors (LP) would like to thank the Natural Sciences and Engineering Research Council Canada for the award of a Visiting Fellowship.

## References

Clark-Lewis, J. W., Edgar, J. A., Shannon, J. S. & Thompson, M. J. (1964). *Aust. J. Chem.* **17**, 877–893.
Clark-Lewis, J. W., Moody, K. & Thompson, M. J. (1970). *Aust. J. Chem.* **23**, 1249–1273.
Coppens, P., Leiserowitz, L. & Rabinovich, D. (1965). *Acta Cryst.* **18**, 1035–1038.
*International Tables for X-ray Crystallography* (1974). Vol. IV, pp. 72–98. Birmingham: Kynoch Press.
Langs, D. A. & DeTitta, G. T. (1975). *Acta Cryst.* A**31**, *S*16.
Larson, A. C. (1970). *Crystallographic Computing*, edited by F. R. Ahmed, pp. 291–294. Copenhagen: Munksgaard.
Larson, A. C. & Gabe, E. J. (1978). *Computing in Crystallography*, edited by H. Schenk, R. Olthof-Hazekamp, H. van Koningsveld & G. C. Gassi, p. 81. Delft Univ. Press.
Matthews, B. W. (1965). *Acta Cryst.* **18**, 151–157.
Quick, A. & Williams, D. J. (1976). *Can. J. Chem.* **54**, 2482–2487.
Rae, A. D. (1965). *Acta Cryst.* **19**, 683–684.
Stewart, R. F., Davidson, E. R. & Simpson, W. T. (1965). *J. Chem. Phys.* **42**, 3175–3187.

---

*Acta Cryst.* (1983). C**39**, 1688–1690

# Structure of 3-Deazauracil (4-Hydroxy-2-pyridone), $C_5H_5NO_2$

By J. N. Low and C. C. Wilson

*Carnegie Laboratory of Physics, University of Dundee, Dundee DD*1 4*HN, Scotland*

(*Received* 24 *May* 1983; *accepted* 9 *August* 1983)

**Abstract.** $M_r = 111{\cdot}1$, $P2_12_12_1$, $a = 8{\cdot}638$ (6), $b = 5{\cdot}279$ (5), $c = 11{\cdot}220$ (8) Å, $U = 511{\cdot}61$ Å$^3$, $Z = 4$, $D_x = 1{\cdot}442$ g cm$^{-3}$, Mo $K\alpha$, $\lambda = 0{\cdot}71069$ Å, $\mu = 0{\cdot}072$ mm$^{-1}$, $F(000) = 232{\cdot}0$, $T = 293$ K, $R = 0{\cdot}065$ for 1133 observed reflections (Friedel pairs not merged). The molecule exists in the 4-enol form. There is an intermolecular N—H···O hydrogen bond.

**Introduction.** In order to make electron spin resonance studies of free radicals produced by ionizing radiations in single crystals, the crystal structure must be known. This structure determination was undertaken as part of a series of such studies which one of us (CCW) is making of nucleic acid constituents.

**Experimental.** Approximately spherical bipyramidal prisms of diameter 0·75 mm from aqueous ethanol. Cell dimensions by diffractometry, Stoe Stadi II diffractometer, crystals mounted along **a** and **b**. Programs *SHELX76* (Sheldrick, 1976), *XANADU* (Roberts & Sheldrick, 1975) and *PLUTO* (Motherwell & Clegg, 1978) used. All calculations performed on the Dundee University DEC-10 computer. Intensities from *a*-axis crystal in the range $h = 0$ to 5, $0° < 2\theta < 55°$, and the *b*-axis crystal in the range $k = 0$ to 5, $0° < 2\theta < 55°$. $(\sin\theta/\lambda)_{\max} = 0{\cdot}77$ Å$^{-1}$; absorption corrections not applied; range of indices: $-12 \le h \le 12$, $0 \le k \le 6$, $0 \le l \le 15$; a standard reflexion measured every 100 reflexions on each layer line revealed a random intensity variation (<5%). 1358 reflexions (*hkl* and $-hkl$), 713 unique, $R_{\rm int} = 0{\cdot}046$, 1133 observed $[F > 5\sigma(F)]$. Structure determined by direct methods; non-hydrogen atoms refined by least squares (on *F*) using anisotropic temperature factors. A difference Fourier map revealed peaks at H-atom distances from N(1), C(3), C(5) and C(6); all H atoms except H(1) included as riding atoms at calculated positions; H(1) fixed at its position as obtained from the difference map; $R = 0{\cdot}065$, $wR = 0{\cdot}120$, $w = 0{\cdot}4459/[\sigma^2(F) + 0{\cdot}029174F^2]$; $(\Delta/\sigma)_{\max} < 0{\cdot}01$; $(\Delta\rho)_{\max} = 0{\cdot}58$, $(\Delta\rho)_{\min} = -0{\cdot}75$ e Å$^{-3}$; scattering factors from *International Tables for X-ray Crystallography* (1974); no correction for secondary extinction.

**Discussion.** The atomic numbering used is shown in Fig. 1, and atomic parameters are listed in Table 1.* Interatomic distances and bond angles are in Table 2. A comparison of these values with those for the deazauracil moiety in the structure of 3-deazauridine (Schwalbe & Saenger, 1973) shows reasonable agreement. As in that structure, the evidence here points strongly in favour of the 4-enol form (see Fig. 1). The presence of H(1) connected to N(1) along with the fact that the C(2)—O(2) bond is a double bond rules out the 2,4-diol form of the molecule, which therefore is confirmed as existing as the 4-enol tautomer. Further, within the pyridine ring there is some electron delocalization over the N(1)—C(2)—C(3)—C(4) moiety, whereas C(5)—C(6) has strong double-bond character [1·348 (4) Å]. The C(4)—O(4) bond is very short compared with similar bonds; however, the short

Table 1. *Atomic coordinates* (×10$^4$) *and equivalent isotropic thermal parameters* (Å$^2$ × 10$^3$), *with e.s.d.'s in parentheses*

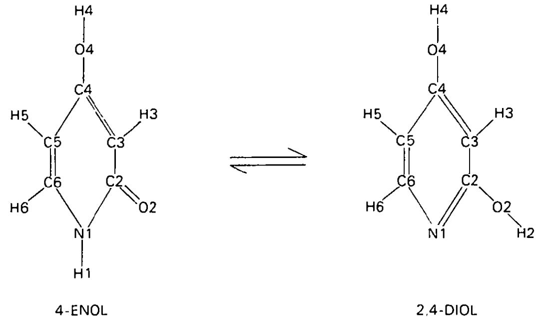
$$U_{\rm eq} = \tfrac{1}{3}\textstyle\sum_i\sum_j U_{ij}a_i^*a_j^*\mathbf{a}_i.\mathbf{a}_j.$$

| | $x$ | $y$ | $z$ | $U_{\rm eq}$ |
|---|---|---|---|---|
| N(1) | 3255 (3) | 3429 (5) | 7102 (2) | 27 (1) |
| H(1) | 4086 | 3167 | 7403 | † |
| C(2) | 3245 (3) | 5441 (5) | 6344 (2) | 26 (1) |
| O(2) | 4417 (3) | 6865 (5) | 6315 (2) | 36 (1) |
| C(3) | 1899 (3) | 5811 (5) | 5650 (2) | 24 (1) |
| C(4) | 671 (3) | 4147 (5) | 5735 (2) | 26 (1) |
| O(4) | −638 (2) | 4432 (5) | 5143 (2) | 37 (1) |
| C(5) | 761 (4) | 2074 (6) | 6526 (3) | 32 (1) |
| C(6) | 2055 (4) | 1774 (6) | 7184 (2) | 32 (1) |

† $U_{\rm iso} = 0{\cdot}05$ Å$^2$.

Table 2. *Interatomic distances* (Å) *and angles* (°)

| | | | |
|---|---|---|---|
| H(1)—N(1) | 0·806 | C(2)—N(1)—H(1) | 113·7 |
| C(2)—N(1) | 1·360 (4) | C(6)—N(1)—H(1) | 122·7 |
| C(6)—N(1) | 1·359 (4) | C(6)—N(1)—C(2) | 122·7 (2) |
| O(2)—C(2) | 1·262 (4) | O(2)—C(2)—N(1) | 118·4 (2) |
| C(3)—C(2) | 1·412 (3) | C(3)—C(2)—N(1) | 117·2 (2) |
| C(4)—C(3) | 1·381 (4) | C(3)—C(2)—O(2) | 124·3 (2) |
| O(4)—C(4) | 1·319 (3) | C(4)—C(3)—C(2) | 120·4 (2) |
| C(5)—C(4) | 1·411 (4) | O(4)—C(4)—C(3) | 123·4 (2) |
| C(6)—C(5) | 1·348 (4) | C(5)—C(4)—C(3) | 119·6 (3) |
| | | C(5)—C(4)—O(4) | 116·9 (3) |
| | | C(6)—C(5)—C(4) | 118·8 (3) |
| | | C(5)—C(6)—N(1) | 121·3 (2) |



4-ENOL 2,4-DIOL

Fig. 1. Atomic numbering of the tautomers.

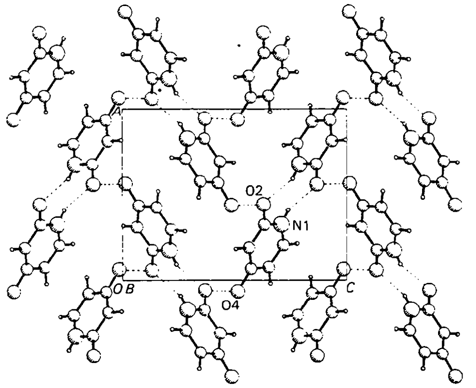


Fig. 2. Contents of the unit cell showing the hydrogen bonding, viewed along **b**.

---

* Lists of structure amplitudes, anisotropic thermal parameters and calculated H-atom parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 38801 (11 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

intermolecular contact of 2·550 (4) Å between O(4) and O(2) (−0·5 + $x$, 1·5 − $y$, 1 − $z$), the latter atom having a deviation of 0·03 (1) Å from the mean plane of the pyridine ring to which O(4) is attached, the C(4)—O(4)···O(2) angle being 113·2 (3)°, strongly suggests the presence of an H atom associated with O(4). All attempts to locate this H atom were unsuccessful. The short intermolecular contact, 2·807 (4) Å, between N(1) and O(2) (1 − $x$, −0·5 + $y$, 1·5 − $z$) is a hydrogen bond, the H(1)···O(2) distance being 2·05 Å, and the N(1)—H(1)···O(2) angle 156°. The pyridine ring in the structure is essentially planar [maximum deviation of an atom from the plane is 0·008 (2) Å for N(1)], another factor in favour of the 4-enol designation (Schwalbe & Saenger, 1973). The contents of the unit cell are shown in Fig. 2.

### References

*International Tables for X-ray Crystallography* (1974). Vol. IV. Birmingham: Kynoch Press.

Motherwell, W. D. S. & Clegg, W. (1978). *PLUTO*. A program for plotting molecular and crystal structures. Univ. of Cambridge, England.

Roberts, P. & Sheldrick, G. M. (1975). *XANADU*. A program for torsion-angle, mean-plane and librational-correction calculations. Univ. of Cambridge, England.

Schwalbe, C. H. & Saenger, W. (1973). *Acta Cryst.* B**29**, 61–69.

Sheldrick, G. M. (1976). *SHELX76*. A program for crystal structure determintion. Univ. of Cambridge, England.

---

*Acta Cryst.* (1983). C**39**, 1690–1692

# Structure of 8-Methoxy-5-oxo-6,8-diazatetracyclo[7.4.0.$0^{2,4}$.$0^{3,7}$]trideca-1(9),10,12-triene-4-carboxylic Acid, $C_{13}H_{12}N_2O_4$

By I. Ueda

*College of General Education, Kyushu University, Ropponmatsu, Chuo-ku, Fukuoka* 810, *Japan*

and Y. Kaku, S. Saeki and M. Hamana

*Faculty of Pharmaceutical Sciences, Kyushu University, Maidashi, Higashi-ku, Fukuoka* 812, *Japan*

(*Received* 31 *May* 1982; *accepted* 1 *August* 1983)

**Abstract.** $M_r = 260·2$, triclinic, $P\bar{1}$, $a = 9·891$ (3), $b = 13·653$ (4), $c = 9·725$ (3) Å, $\alpha = 105·13$ (3), $\beta = 110·23$ (2), $\gamma = 75·95$ (2)°, $V = 1172·6$ (6) Å$^3$, $Z = 4$, $D_m = 1·470$, $D_x = 1·422$ Mg m$^{-3}$, $\lambda$(Mo $K\alpha$) = 0·71069 Å, $\mu = 0·12$ mm$^{-1}$, $F(000) = 544$, $T = 293$ K. Final $R = 0·048$ for 4686 observed reflexions. The condensed quinolinium and lactam rings form a hinge in which the three-membered ring is inserted. The carbonyl and carboxy moieties form one plane with the lactam ring *via* the intramolecular O···HO hydrogen bond. A tetramer is formed by the two paired intermolecular O···HN hydrogen bonds in the crystal.

**Introduction.** In the course of studies on the reactions of quinolinium salts with active methylene compounds, reaction of *N*-methoxyquinolinium perchlorate with methyl bromocyanoacetate was found to afford the title compound ($E$) in the yield 25·5% as shown in Fig. 1. The formation of $E$ may be rationalized by the course shown also in Fig. 1. Initially, the nucleophilic attack of a carbanion of $B$ on $A$ gives a 1,4-dihydroquinoline ($C$). The next step is the transformation of $C$ to a cyclopropane ring compound ($D$) by nucleophilic attack of the 3-position at the side-chain C with the concerted elimination of the Br$^-$ ion. Further, a C—N bond is formed between the quinolinium C and the N atom of the cyano group in $D$ to produce the title product $E$ after several steps. By an X-ray diffraction method the structure of $E$ was determined as given in the title. The molecular structure resembles that of benzomorphans which have analgetic and antagonistic activities; however, derivatives of the title compound have little activity.

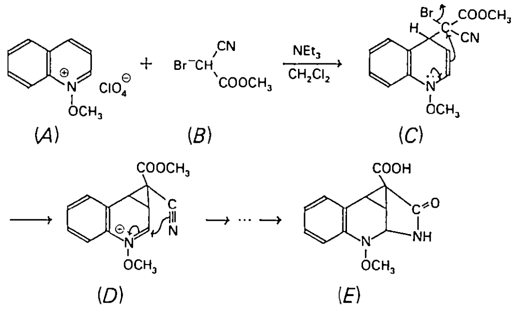


Fig. 1. Reaction scheme.