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Structure of a Novel and Reproducible Polymorph (*Z*) of the Histamine H_2-Receptor Antagonist Cimetidine, $C_{10}H_{16}N_6S$

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Abstract. $M_r = 252{\cdot}34$, $P2_1/c$, $a = 7{\cdot}283\ (2)$, $b = 10{\cdot}808\ (2)$, $c = 18{\cdot}281\ (3)$ Å, $\beta = 118{\cdot}18\ (2)°$, $V = 1268{\cdot}4\ (5)$ Å^3, $D_x = 1{\cdot}321$ Mg m^{-3}, $Z = 4$, Mo $K\alpha$, $\lambda = 0{\cdot}71073$ Å, $\mu = 0{\cdot}24$ mm^{-1}, $F(000) = 536$, $T = 295\ (2)$ K, $R = 0{\cdot}034$ for 1733 unique reflexions. The crystal structure of a novel polymorph (*Z*) of cimetidine is compared with that of *A* reported by Hädicke, Frickel & Franke [*Chem. Ber.* (1978), **111**, 3222–3232]. Its bond distances and angles (except three angles) agree well with the corresponding ones observed in polymorph *A*. However, due to the full rearrangement (*i.e.* geometrical isomerization) of the guanidine moiety, their conformations differ markedly; *e.g.* the intramolecular bond distance N(2)···N(4) = 2·881 (2) Å of form *A*, which corresponds to a strong hydrogen bond, increased to 7·382 (3) Å. Consequently, the hydrogen bonding of *Z* also differs significantly from that of *A*.

Introduction. The efficacious histamine H_2-receptor antagonist cimetidine [*N*-cyano-*N'*-methyl-*N''*-(2-{[(5-methyl-1*H*-imidazol-4-yl)methyl]thio}ethyl)guanidine] is known to exhibit rich polymorphism, *e.g.* three crystal modifications are mentioned by Bavin, Sly, Tovey & Ward (1976). The existence of further polymorphs is assumed by Kojić-Prodić, Kajfez, Belin, Toso & Sunjić (1979) on the basis of IR spectra and X-ray powder diagrams. However, owing to the coprecipitation of these polymorphs neither recipes for their reproducible preparations, nor descriptions of their structures could be provided. Only the crystal structure of the modification already used in human pharmacotherapy (hereinafter polymorph *A*) has been reported so far (Hädicke, Frickel & Franke, 1978). Consequently, it is of primary importance to describe the preparation and the crystal structure of a novel cimetidine polymorph termed *Z* which in contrast to the other hardly reproducible ones can be prepared from aqueous solution in unique form. Since the sand-like *Z* form of cimetidine adsorbs only a low amount of water it is a rather profitable intermediate in the industrial production of the registered form *A*.

Experimental. Crystals of polymorph *Z* prepared as follows: 20 g cimetidine mixed with 40% (v/v) methanol/water solution; from the well stirred suspension a homogenous solution was obtained by adding 7 ml of CH_3COOH; after cleaning and filtering this solution cimetidine was precipitated with NH_4OH at 283 K (pH = 9); crystals were filtered and dried; m.p. 414–416 K. Crystal $0{\cdot}1 \times 0{\cdot}12 \times 0{\cdot}22$ mm. Enraf–Nonius CAD-4 diffractometer, graphite monochromator. Cell constants by least squares using 25 reflexions, $20° \le 2\theta \le 30°$. Systematic absences *h*

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$= 2n + 1$ in $h0l$, $k = 2n + 1$ in $0k0$. $2\theta_{max} = 50°$; $h\,0\rightarrow8$, $k\,0\rightarrow12$, $l\,\bar{2}1\rightarrow18$. Three standard reflexions (0,4,10, 2,3,$\bar{12}$, 3,4,$\bar{12}$), average max. variation 7·8%. 2228 measured reflexions, 1733 unique, 485 unobserved [$I < 3{\cdot}0\sigma(I)$]. No absorption correction. Structure solved by *MULTAN*78 (Main, Hull, Lessinger, Germain, Declercq & Woolfson, 1978) using reflexions with $E \geq 1{\cdot}40$. Full-matrix least squares, $\sum w(\Delta F)^2$ minimized, 155 parameters refined. Final $R = 0{\cdot}034$, $R_w = 0{\cdot}042$, $S = 2{\cdot}23$, $w = [\sigma^2(F_o) + 0{\cdot}25 \times 10^{-4}F_o^2]^{-1}$. $(\Delta/\sigma)_{max}$ 0·081. Max. and min. peak heights in final $\Delta\rho$ map $\pm$ 0·29 e Å^{-3}. Extinction coefficient 0·81707 $\times$ 10^{-6}. Positions of H atoms bound to C atoms were generated from assumed geometries; those belonging to N atoms were located in a difference Fourier synthesis at $R = 0{\cdot}115$; they were treated isotropically in the final stage of the refinement. Scattering factors from *International Tables for X-ray Crystallography* (1962). Program system applied: Enraf–Nonius *SDP*-34 with local modifications adapted to a PDP 11/34 minicomputer (64 K). The identical crystal form of the single crystals and the sand-like powder of cimetidine was checked from an X-ray powder diagram by comparing it with a theoretical pattern computed from the atomic coordinates listed in Table 1* using the program *LAZY PULVERIX* (Yvon, Jeitschko & Parthé, 1977).

Discussion. The bond distances and the majority of the bond angles assumed by the cimetidine molecule in form *Z* (Table 2) agree well with the corresponding ones observed in form *A*; the r.m.s. of the 17 bond-length discrepancies is only 0·008 Å, while that for 17 bond angles is 0·7°. This corresponds to an agreement between two independent structure analyses of a crystal of medium quality. However, there are three bond angles [S(1)–C(6)–C(7), N(3)–C(8)–N(5) and N(4)–C(8)–N(5)] which show significant differences. These may be attributed to the markedly different conformations of the cimetidine molecule assumed in the *A* and *Z* polymorphs (Fig. 1). In conformer *A*, characterized by a ten-membered ring closed by an intramolecular hydrogen bond N(4)–H(N4)···N(2), the quasi-collinear N(5)–C(10)–N(6) moiety and the N(4)-methyl group point outward from the macro-ring keeping themselves separated optimally by attaining the geometrical isomer *E* (*anti*) across the C(8)–N(5) double bond. Consequently, the N(5)–C(10) and C(8)–N(3) multiple bonds are eclipsed and their repulsion opens the N(3)–C(8)–N(5) angle at the expense of the adjacent N(4)–C(8)–N(5). In contrast to this the planar guanidine moiety in conformer *Z*

* Lists of structure factors, anisotropic thermal parameters and H-atom parameters have been deposited with the British Library Lending Division as Supplementary Publication No. SUP 39017 (13 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 1. *Final fractional atomic coordinates* ($\times 10^4$) *for non-hydrogen atoms and equivalent isotropic temperature parameters with e.s.d.'s in parentheses*

$B_{eq} = 4[(b_{11}b_{22}b_{33})/(a^{*2}b^{*2}c^{*2})]^{1/3}$.

	x	*y*	*z*	B_{eq}(Å^2)
S(1)	198 (1)	2499 (1)	2605 (1)	3·15 (1)
N(1)	−36 (2)	5764 (1)	775 (1)	2·52 (4)
N(2)	−1499 (2)	4041 (1)	887 (1)	2·82 (4)
N(3)	2541 (2)	3960 (1)	4354 (1)	2·26 (3)
N(4)	5307 (2)	2879 (1)	5349 (1)	2·42 (3)
N(5)	5406 (2)	3663 (1)	4168 (1)	2·46 (4)
N(6)	9137 (2)	3237 (2)	4714 (1)	2·92 (4)
C(1)	−1799 (3)	5093 (2)	493 (1)	2·99 (5)
C(2)	599 (2)	4034 (2)	1463 (1)	2·27 (4)
C(3)	1514 (2)	5099 (2)	1400 (1)	2·35 (4)
C(4)	3680 (3)	5574 (2)	1861 (1)	4·06 (6)
C(5)	1575 (3)	2992 (2)	2046 (1)	2·93 (5)
C(6)	−392 (2)	3956 (2)	2927 (1)	2·96 (5)
C(7)	1445 (2)	4654 (2)	3587 (1)	2·59 (4)
C(8)	4446 (2)	3486 (2)	4632 (1)	1·97 (4)
C(9)	4419 (2)	2813 (2)	5910 (1)	2·93 (5)
C(10)	7389 (2)	3413 (2)	4492 (1)	2·07 (4)

Table 2. *Bond lengths* (Å), *bond angles* (°) *and their differences* Δ *from the corresponding values observed in conformer A (Hädicke et al.,* 1978)

The e.s.d.'s are given in parentheses.

		Δ ($\times 10^2$Å)			Δ ($\times 10^2$Å)
S(1)–C(5)	1·817 (2)	1·1	C(3)–C(4)	1·485 (2)	−0·1
S(1)–C(6)	1·802 (2)	1·2	N(3)–C(8)	1·334 (2)	0
N(1)–C(1)	1·347 (2)	−1·5	N(4)–C(8)	1·329 (2)	0·1
N(1)–C(3)	1·370 (2)	0·4	N(4)–C(9)	1·450 (2)	0
N(2)–C(1)	1·308 (2)	0·3	N(5)–C(8)	1·344 (2)	0·2
N(2)–C(2)	1·387 (2)	−0·1	N(5)–C(10)	1·304 (2)	1·5
N(3)–C(7)	1·452 (2)	0·9	N(6)–C(10)	1·155 (2)	−0·1
C(2)–C(3)	1·362 (3)	0·1	C(6)–C(7)	1·513 (2)	−0·5
C(2)–C(5)	1·481 (3)	1·3			

		Δ(°)			Δ(°)
C(5)–S(1)–C(6)	102·0 (2)	0·9	N(1)–C(3)–C(2)	105·7 (2)	−0·9
C(1)–N(1)–C(3)	107·3 (2)	0·8	N(1)–C(3)–C(4)	121·8 (2)	0·5
C(1)–N(2)–C(2)	105·1 (2)	−0·4	C(2)–C(3)–C(4)	132·5 (3)	0·4
C(7)–N(3)–C(8)	124·4 (2)	1·0	S(1)–C(5)–C(2)	115·0 (2)	0·7
C(8)–N(4)–C(9)	124·2 (2)	−0·3	S(1)–C(6)–C(7)	116·0 (2)	−3·7
C(8)–N(5)–C(10)	118·6 (2)	0·1	N(3)–C(7)–C(6)	112·8 (2)	0·3
N(1)–C(1)–N(2)	112·1 (2)	0·1	N(3)–C(8)–N(4)	119·1 (2)	0·8
N(2)–C(2)–C(3)	109·8 (2)	0·4	N(3)–C(8)–N(5)	117·5 (2)	5·6
N(2)–C(2)–C(5)	122·2 (2)	−1·6	N(4)–C(8)–N(5)	123·3 (2)	−6·3
C(3)–C(2)–C(5)	128·0 (3)	1·2	N(5)–C(10)–N(6)	173·7 (3)	−0·3

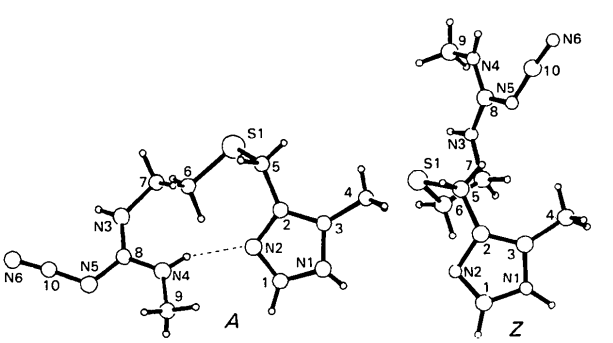

Fig. 1. Perspective views of conformers *A* (Hädicke *et al.*, 1978) and *Z* with atomic numbering. Bare numbers are for C unless indicated otherwise. The H atoms are shown but not labelled.

Table 3. *Relevant torsion angles of conformer Z together with their differences Δ from the corresponding angles observed in conformer A (Hädicke et al., 1978)*

The e.s.d.'s are given in parentheses.

	$\varphi(Z)$	Δ
N(2)–C(2)–C(5)–S(1)	−49·4 (2)°	−20·4°
C(2)–C(5)–S(1)–C(6)	−45·2 (2)	107·0
C(5)–S(1)–C(6)–C(7)	−69·8 (2)	132·5
S(1)–C(6)–C(7)–N(3)	−60·1 (2)	−115·6
C(6)–C(7)–N(3)–C(8)	110·1 (3)	−19·5
C(7)–N(3)–C(8)–N(4)	179·2 (3)	−188·1
C(7)–N(3)–C(8)–N(5)	−0·1 (2)	172·6
N(3)–C(8)–N(4)–C(9)	−7·6 (2)	178·7
N(3)–C(8)–N(5)–C(10)	167·6 (3)	−180·1

practically undergoes a full rearrangement by a *ca* 180° rotation about each of its three C–N bonds. Thus the N(5)–C(10) and C(8)–N(4) multiple bonds are eclipsed forcing these two bond angles into the opposite directions as follows:

	A		*Z*
N(3)–C(8)–N(5)	123·1 (2)°	→	117·5 (2)°
N(4)–C(8)–N(5)	117·0 (2)	→	123·3 (2).

Namely, in conformer *Z* the geometrical isomer *Z* (*syn*) is the preferred one since the other isomer would bring the cyano moiety too close to the methylene group of C(7). It is worth noting that neither of these two arrangements of the N(5)–C(10)–N(6) group could exist within the other conformer. The simultaneous rotations about the C–N bonds of the planar guanidine moiety should occur, however, against the delocalized $p\pi$ multiple bonds of the six-atom system, presumably at the expense of solvation energies. Beyond this rearrangement of the *N*-cyano-*N'*-methylguanidine moiety observed also in the form of cimetidine monohydrate (Kojić-Prodić, Ružić-Toroš, Bresciani-Pahor & Randaccio, 1980) the title compound, as expected by Lipinski (1983), possesses great flexibility about the four-atom chain binding the imidazole ring and the guanidine moiety. In the novel *Z* conformation the ten-membered ring disappears and the side chain becomes elongated making a semi-helical turn about an axis bisecting N(2) and N(4) atoms separated now at a distance of 7·382 (3) Å. [Note that the N(2)···N(4) distance in form *A* is 2·881 (2) Å.] This helical turn may also account for the increase of the third bond angle S(1)–C(6)–C(7) by 3·7°. These alterations in the conformation are shown by the torsion-angle differences $\Delta = \varphi(A) - \varphi(Z)$ listed together with the relevant torsion angles for conformer *Z* in Table 3. The greatest rotation is observed about the S(1)–C(6) bond; the second greatest occurs at the adjacent C(6)–C(7) bond, but in the opposite direction. The great pliability of this four-atom chain is also demonstrated by the torsion angles found in the crystal of cimetidine monohydrate by Kojić-Prodić *et al.* (1980). In particular, the amount of rotation about the S(1)–C(6) and C(7)–N(3) bonds [$\varphi_{SC} = 137{\cdot}2$ (2) and $\varphi_{CN} = -75{\cdot}8$ (4)°] differs from the corresponding ones in both *A* and *Z* conformers resulting in a basically different molecular shape.

The analysis of the hydrogen-bond networks observed in these two polymorphs suggests that the three NH···N hydrogen bonds in each play an important role in their formation and presumably in those polymorphs whose existence has been assumed in the literature (Bavin *et al.*, 1976; Kojić-Prodić *et al.*, 1979). The most intriguing result is that the intramolecular N(4)–H(N4)···N(2) linkage is replaced by an energetically more favourable [*cf.* cooperative effects (Del Bene & Pople, 1973; Hinton & Harpool, 1977)] intermolecular N(4)–H(N4)···N(2) bond formed between the glide-plane-related molecules (Fig. 2). The second hydrogen bond of form *A* forming dimers around centres of symmetry in form *Z* binds infinite chains of molecules along the **a** axis. Consequently, it is also more favourable than the isolated cyclic dimeric form. The third active H atom linked to the imidazole ring has to change its acceptor, however, from N(5) of the glide-plane-related molecules in polymorph *A* to N(6) which builds up infinite helices around the screw axes 2_1 ($\frac{1}{2}$, *y*, $\frac{1}{4}$). Thus N(6) acts as a bifurcated acceptor in polymorph *Z*.

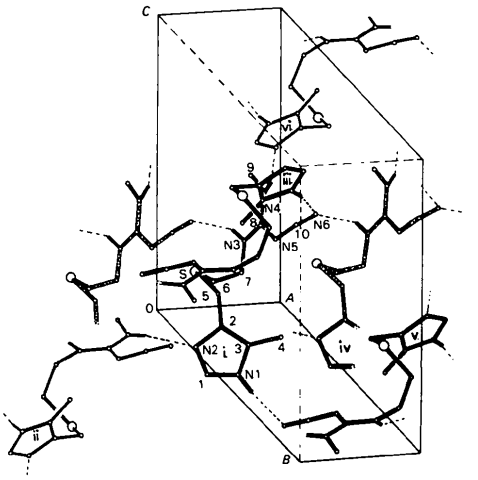

Fig. 2. Intermolecular hydrogen bonding of polymorph *Z*. Irrelevant H atoms are omitted. Hydrogen-bond parameters (Å and deg):

	N···N	H···N	∠NH···N
N(2^{i})···H(N4)–N(4^{ii})	2·919 (2)	2·08 (2)	160·2 (20)
N(6^{i})···H(N1)–N(1^{iii})	2·988 (2)	2·06 (2)	171·6 (18)
N(6^{i})···H(N3)–N(3^{iv})	2·956 (2)	2·10 (2)	154·5 (18).

Symmetry code: (i) *x*, *y*, *z*; (ii) *x*−1, 0·5−*y*, −0·5+*z*; (iii) 1−*x*, −0·5+*y*, 0·5−*z*; (iv) 1+*x*, *y*, *z*; (v) 1−*x*, 0·5+*y*, 0·5−*z*; (vi) 1+*x*, 0·5−*y*, 0·5+*z*.

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Stereochemical Studies of Oligomers. IX. * Structures of 1,4-Butanediyl Bis[*o*-(*m*- and *p*-)chlorobenzoates] (BDDO, BDDM and BDDP), $C_{18}H_{16}Cl_2O_4$

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Abstract. $M_r = 367{\cdot}2$, room temperature, Ni-filtered Cu $K\alpha$ radiation, $\lambda = 1{\cdot}5418$ Å. BDDO: monoclinic, $P2_1/n$, $a = 8{\cdot}858$ (1), $b = 12{\cdot}956$ (2), $c = 7{\cdot}472$ (1) Å, $\beta = 97{\cdot}08$ (2)°, $Z = 2$, $V = 850{\cdot}98$ (20) Å^3, $D_x = 1{\cdot}44$ g cm^{-3}, $\mu = 36{\cdot}6$ cm^{-1}, $F(000) = 380$, $R = 0{\cdot}050$, 1183 observed reflections. BDDM: monoclinic, $P2_1/c$, $a = 5{\cdot}870$ (3), $b = 19{\cdot}270$ (2), $c = 7{\cdot}651$ (2) Å, $\beta = 97{\cdot}29$ (3)°, $Z = 2$, $V = 858{\cdot}45$ (50) Å^3, $D_x = 1{\cdot}42$ g cm^{-3}, $\mu = 36{\cdot}3$ cm^{-1}, $F(000) = 380$, $R = 0{\cdot}049$, 1301 observed reflections. BDDP: triclinic, $P1$, $a = 7{\cdot}609$ (2), $b = 10{\cdot}318$ (1), $c = 5{\cdot}924$ (3) Å, $\alpha = 96{\cdot}31$ (2), $\beta = 98{\cdot}35$ (3), $\gamma = 111{\cdot}05$ (2)°, $Z = 1$, $V = 422{\cdot}79$ (26) Å^3, $D_x = 1{\cdot}44$ g cm^{-3}, $\mu = 36{\cdot}8$ cm^{-1}, $F(000) = 190$, $R = 0{\cdot}081$, 1006 observed reflections. The usual tilt of the carboxylic groups with respect to their phenyl rings is observed and in the case of the *ortho* derivative the steric hindrance increases the value [31·2 (1)°] noticeably. The conformation of the butanediyl chain is *gauche–trans–gauche* in the *ortho* derivative and all-*trans* for the other two compounds.

Introduction. During studies on polymers in the late nineteen forties, it was often found that stereochemistry strongly influences polymerization mechanisms and relationships between molecular structure and physical and mechanical properties of polymers. In particular, a review of experiments on polyester derivatives made over the course of a decade at the University of Montreal (Brisse, Marchessault & Pérez, 1980) shows that substantial progress in understanding their structures was made if their stereochemistry was absolutely certain. For this class of polymers the polymeric chain may be symbolically divided into a number of monomers and the knowledge of the structure of these small 'model compounds' is essential if the correct polymer structure is to be obtained. This approach was employed in the structural determination of a large number of biopolymers (Brant, Tonelli & Flory, 1969; Pérez & Brisse, 1978; Pérez & Marchessault, 1978) or polymers (Benedetti, Pedone & Allegra, 1970) and to confirm the structure of poly(ethylene terephthalate) on the basis of those of ethylene glycol dibenzoate (Pérez & Brisse, 1976*a*) and of its *p*-chloro derivative (Pérez & Brisse, 1975). Moreover, the possible structural conformation of the not yet known poly(trimethylene terephthalate) was hypothesized on the basis of that of related monomers: trimethylene glycol dibenzoate (Pérez & Brisse, 1977) and its *p*-chloro derivative (Pérez & Brisse, 1976*b*). From these examples it is clear that structural data on monomers provide a powerful basis for investigating the structures of macromolecules. Consequently, increasing attention is devoted to the conformations of these types of monomers in order to obtain precise geometrical information which is also useful in the energy calculations.

* Part VIII: Bocelli & Grenier-Loustalot (1983*c*).

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