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Hydrogen-bonding in organosilanediols. The crystal structure of bis(pentamethylcyclopentadienyl)silanediol

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Abstract

The unit cell of $(C_5Me_5)_2Si(OH)_2$ contains twelve molecules arranged as three cyclic tetramers formed by hydrogen-bonding. One tetramer has a centre of symmetry while the other two are unsymmetrical, giving six independent molecules in the unit cell. There is no hydrogen-bonding between tetramers.

Introduction

Organosilanediols $R_2Si(OH)_2$ can form a wide variety of hydrogen-bonded crystal structures. When R is relatively small, complicated infinite hydrogen-bonded structures are formed; thus $Et_2Si(OH)_2$ has hydrogen-bonded chains cross-linked to form a layer [1], while $Ph_2Si(OH)_2$ has hydrogen-bonded hexamers which are further linked by hydrogen bonds to form an infinite three-dimensional network [2]. For the diols $R_2Si(OH)_2$ in which R is a more bulky group, namely i-Pr [3], t-Bu [4], C_6H_{11} (cyclohexyl) [5], or *o*-tolyl [6]), the hydrogen-bonding results in cyclic dimers which are further linked to form chains, as shown diagrammatically (I). In contrast, the diol $(Me_3Si)_3CSiPh(OH)_2$, which contains the very bulky $(Me_3Si)_3C$ group, forms discrete cyclic dimers [7]. In view of the wide range of possible structural types, when the diol $(C_5Me_5)_2Si(OH)_2$ became available at the University of Bielefeld it seemed of interest to determine its structure. The results presented below show that it forms yet another type of hydrogen-bonded structure in the crystal.

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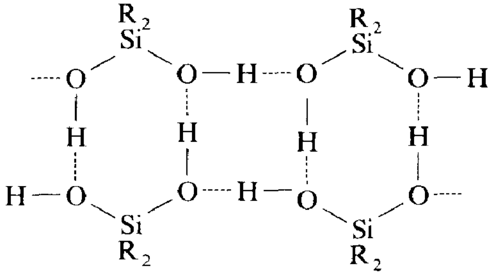

(I)

Experimental

The pyridine-*N*-oxide (Aldrich) was used as supplied, and evidently contained some moisture.

$(C_5Me_5)_2Si(OH)_2$

A solution of the pyridine-*N*-oxide (0.60 g, 6.3 mmol) in toluene (10 cm^3) was added dropwise to a solution of $(\eta^5\text{-}C_5Me_5)_2Si$ [8] (1.70 g, 5.7 mmol) in toluene (18 cm^3) at −60 °C. The solution was allowed to warm to room temperature during 16 h then volatile materials were removed under vacuum to leave a brown residue, from which $(C_5Me_5)_2Si(OH)_2$ was sublimed (7.6 mmHg, 90 °C) as a colourless solid (0.795 g, 42%), m.p. 123–125 °C; (Found: C, 72.62; H, 9.48. $C_{20}H_{32}O_2Si$ calcd.: C, 72.23; H, 9.70%). δ(H) (300.13 MHz, $CDCl_3$): 1.03, 1.74, 1.78 (s, 6H : 12H : 12H, $(C_5(CH_3)_5)$), 2.16 (s, 2H, $Si(OH)_2$); δ(C) (75.43 MHz, $CDCl_3$): 11.4, 12.0, 15.7 $((C_5(CH_3)_5)$, 54.8 (allyl-$C_5(CH_3)_5$), 135.6, 138.2 (vinyl-$C_5(CH_3)_5$); δ(Si) NMR (53.63 MHz, $CDCl_3$), −18.8; *m/z* (70 eV); 332 (14% $[M]^+$), 197 (100 $[M - C_5Me_5]^+$), 179 (17 $[C_5Me_5SiO]^+$). Crystals suitable for X-ray crystallography were obtained from hexane. A crystal of dimensions 0.37 × 0.35 × 0.37 mm, sealed in a thin-walled capillary, was used for the structure determination.

Crystal data. $C_{20}H_{32}O_2Si$, M 332.56, triclinic, space group $P\bar{1}$, a 15.007(4), b 17.048(5), c 24.422(7) Å), α 88.43(2), β 74.13(2), γ 89.55(2)°, U 6009.8 Å^3, $Z = 12$, D_c 1.11 g cm^{-3}, $F(000) = 2184$, monochromated Mo-K_α radiation, λ 0.71069 Å, μ 1.2 cm^{-1}.

Data were measured on an Enraf–Nonius CAD4 diffractometer. Intensities for 8515 unique reflections with $h \pm k \pm l$ and $2 < \theta < 20°$ were measured by a $\theta/2\theta$ scan with a scan width $\Delta\theta = (0.8 + 0.35 \tan\theta)°$. After correction for Lorentz and polarization effects but not for absorption, 3716 reflections with $|F|^2 > 2\sigma(F^2)$ were used for the structure analysis, where $\sigma(F^2) = [\sigma^2(I) + (0.04I)^2]^{1/2}/\text{Lp}$.

The direct methods routines of the SHELXS-program were used to find Si, O, and the carbon atoms bonded to Si, the rest of the atoms were located on difference maps. All the atoms were refined by large block least-squares with isotropic temperature factors using SHELX-76. The hydrogen atoms were omitted. An attempt to refine the carbon atom positions freely resulted in unacceptable geometry for the rings, presumably because of the rather weak and limited data, and so the C–C bonds of the rings and the ring-C–Methyl bonds were set equal to free variables which refined to values of 1.50(3) Å and 1.57(3) Å respectively.

Table 1. Fractional atomic Coordinates($\times 10^4$) with estimated standard deviations in parentheses

	x	*y*	*z*		*x*	*y*	*z*
Si(1)	3269(6)	7537(5)	5140(4)	C(37)	3509(22)	2372(15)	4650(15)
Si(2)	3578(6)	4304(5)	5105(4)	C(38)	1355(20)	2323(14)	4617(14)
Si(3)	4388(6)	3669(5)	8499(4)	C(39)	692(17)	4077(23)	4366(17)
Si(4)	4791(6)	419(5)	8294(4)	C(40)	2435(30)	5323(15)	4129(18)
Si(5)	7516(6)	1834(5)	8287(4)	C(41)	4377(14)	4225(12)	7844(11)
Si(6)	7891(6)	−1438(5)	8102(4)	C(42)	3424(15)	4471(13)	7824(13)
O(1)	2825(12)	6655(11)	5151(8)	C(43)	3336(18)	5346(13)	7771(16)
O(2)	4374(13)	7398(11)	5088(8)	C(44)	4252(17)	5639(13)	7819(14)
O(3)	3782(13)	5266(11)	4966(8)	C(45)	4923(14)	4972(13)	7782(13)
O(4)	4635(14)	3963(12)	5054(9)	C(46)	4847(21)	3758(18)	7295(11)
O(5)	3993(13)	2785(11)	8434(8)	C(47)	2599(18)	3901(16)	7843(13)
O(6)	5480(13)	3543(11)	8474(8)	C(48)	2414(19)	5811(20)	7807(16)
O(7)	4925(13)	1386(11)	8333(8)	C(49)	4491(24)	6530(14)	7677(15)
O(8)	5829(12)	60(10)	8323(8)	C(50)	5993(14)	5130(19)	7630(14)
O(9)	6543(11)	2189(9)	8178(7)	C(51)	3775(16)	4038(15)	9236(13)
O(10)	7361(12)	856(11)	8277(8)	C(52)	4023(18)	4887(15)	9250(18)
O(11)	8309(13)	−553(11)	8181(8)	C(53)	3119(17)	5307(15)	9310(15)
O(12)	6819(14)	−1284(12)	8055(9)	C(54)	2309(17)	4781(15)	9341(15)
C(1)	2734(14)	7943(12)	5877(11)	C(55)	2750(16)	3984(15)	9311(15)
C(2)	3306(15)	8640(12)	5930(13)	C(56)	4112(24)	3521(19)	9682(14)
C(3)	2706(16)	9358(12)	5979(14)	C(57)	5000(19)	5213(21)	9247(16)
C(4)	1793(16)	9136(11)	5887(13)	C(58)	2991(30)	6219(15)	9365(18)
C(5)	1790(14)	8255(11)	5893(13)	C(59)	1258(18)	4986(25)	9429(19)
C(6)	2679(21)	7293(16)	6353(11)	C(60)	2099(21)	3248(16)	9399(15)
C(7)	4296(15)	8655(16)	6030(12)	C(61)	3890(16)	97(14)	9015(12)
C(8)	3050(22)	10230(13)	5946(14)	C(62)	3872(18)	−782(14)	9065(16)
C(9)	980(19)	9735(17)	5922(15)	C(63)	2915(17)	−1007(13)	9048(14)
C(10)	916(15)	7763(15)	5879(12)	C(64)	2291(15)	−304(14)	9105(16)
C(11)	3170(14)	8076(11)	4470(10)	C(65)	2962(15)	368(13)	8963(13)
C(12)	2197(15)	8114(13)	4417(14)	C(66)	4284(24)	394(20)	9506(13)
C(13)	1860(18)	8948(13)	4456(15)	C(67)	4658(23)	−1408(20)	9031(18)
C(14)	2728(17)	9426(14)	4355(16)	C(68)	2434(22)	−1833(15)	9168(14)
C(15)	3485(15)	8911(11)	4459(12)	C(69)	1245(16)	−231(20)	9111(15)
C(16)	3774(21)	7668(18)	3924(11)	C(70)	2636(21)	1248(13)	9023(14)
C(17)	1602(19)	7376(14)	4376(13)	C(71)	4631(18)	217(13)	7559(13)
C(18)	956(21)	9240(23)	4307(17)	C(72)	3779(19)	650(15)	7511(15)
C(19)	2731(25)	10345(14)	4387(16)	C(73)	3005(19)	100(16)	7507(19)
C(20)	4466(15)	9196(17)	4484(13)	C(74)	3351(16)	−707(16)	7606(15)
C(21)	2797(15)	4153(13)	5831(12)	C(75)	4368(16)	−628(13)	7549(13)
C(22)	1811(14)	4355(12)	5857(12)	C(76)	5545(23)	380(25)	7073(16)
C(23)	1261(15)	3613(13)	5902(14)	C(77)	3862(27)	1559(16)	7386(17)
C(24)	1825(17)	2983(14)	6090(14)	C(78)	1977(19)	331(23)	7539(17)
C(25)	2779(16)	3306(14)	6018(14)	C(79)	2809(28)	−1482(19)	7577(19)
C(26)	3129(24)	4656(19)	6267(13)	C(80)	4965(24)	−1379(17)	7586(17)
C(27)	1440(21)	5196(13)	5756(13)	C(81)	8488(16)	2109(13)	7605(11)
C(28)	176(15)	3596(18)	6034(14)	C(82)	9380(17)	1838(12)	7713(14)
C(29)	1466(23)	2120(14)	6240(15)	C(83)	9934(17)	2557(14)	7743(14)
C(30)	3587(21)	2802(20)	6140(16)	C(84)	9428(19)	3266(14)	7606(16)
C(31)	3303(15)	3928(12)	4406(11)	C(85)	8536(18)	2984(13)	7514(14)
C(32)	2927(15)	3116(12)	4572(12)	C(86)	8335(23)	1660(19)	7086(12)
C(33)	1955(16)	3091(13)	4519(15)	C(87)	9732(20)	997(13)	7832(13)
C(34)	1715(16)	3919(13)	4387(13)	C(88)	10942(18)	2600(22)	7814(16)
C(35)	2543(17)	4442(15)	4306(17)	C(89)	9784(26)	4137(15)	7544(16)
C(36)	4194(21)	4011(24)	3891(13)	C(90)	7790(20)	3512(18)	7340(14)

Table 1 (continued)

	x	y	z		x	y	z
C(91)	7582(16)	2080(12)	9028(12)	C(106)	8696(23)	−1118(17)	6892(12)
C(92)	8342(19)	1574(14)	9137(15)	C(107)	10300(20)	−1541(20)	7504(16)
C(93)	9045(19)	2077(13)	9296(14)	C(108)	10293(22)	−3504(19)	7501(16)
C(94)	8836(16)	2896(13)	9125(13)	C(109)	8381(25)	−4060(16)	7168(16)
C(95)	7909(16)	2914(12)	9003(13)	C(110)	7199(16)	−2537(18)	7030(13)
C(96)	6594(18)	1992(21)	9457(14)	C(111)	7806(14)	−2005(12)	8792(11)
C(97)	8219(24)	696(15)	9357(14)	C(112)	7526(17)	−2836(12)	8737(14)
C(98)	9981(19)	1813(21)	9415(16)	C(113)	8272(15)	−3352(12)	8855(11)
C(99)	9411(23)	3633(16)	9197(16)	C(114)	9035(17)	−2854(13)	8947(14)
C(100)	7427(20)	3696(14)	8883(13)	C(115)	8758(16)	−2016(13)	8881(14)
C(101)	8560(15)	−1793(11)	7358(11)	C(116)	7087(20)	−1590(19)	9291(12)
C(102)	9468(16)	−2056(13)	7447(15)	C(117)	6595(17)	−3131(20)	8638(15)
C(103)	9556(16)	−2925(13)	7366(13)	C(118)	8270(27)	−4274(12)	8859(17)
C(104)	8629(17)	−3230(13)	7348(15)	C(119)	9869(21)	−3280(20)	9101(16)
C(105)	8121(16)	−2520(12)	7216(12)	C(120)	9328(22)	−1370(17)	9083(15)

Refinement converged at $R = 0.164$, $R' = 0.162$ with weighting scheme $w = 1/\sigma^2(F)$, and final shift to error ratio < 1.2. A final difference map had peaks of up to 0.6 $e\text{Å}^{-3}$. The high R factor is a consequence of the weak and limited data.

Final atom coordinates are listed in Table 1 and lists of temperature factors, structure factors, and full lists of bond lengths and angles are available from P.B.H.

Results and discussion

The diol $(C_5Me_5)_2Si(OH)_2$ was formed upon treatment of the compound $(\eta^5\text{-}C_5Me_5)_2Si$ [8] with moist pyridine-*N*-oxide. Presumably the compound $(C_5Me_5)_2\text{-}Si{=}O$ is initially formed but reacts readily with water to generate the silanediol.

The high R factor mentioned above means that detailed discussion of bond angles and lengths is not possible; there is no doubt, however, about the molecular structure itself. The hydrogen-bonded structure is clearly different from those of other silanediols in that it contains discrete tetramers. The structures, together with the atom numbering scheme and the hydrogen-bonding, are shown for the symmetrical tetramer in Fig. 1 and the unsymmetrical tetramer in Fig. 2. The unit cell is shown in Fig. 3, where carbon atoms other than those directly attached to the Si atoms are omitted. The tetramer at the centre of the unit cell lies on a crystallographic inversion centre which also relates the other two tetramers which have no imposed symmetry, giving a total of six different independent molecules (labeled **A**–**F** in Figs. 1 and 2) within the unit cell. There appears to be no significant difference between the geometries of the two crystallographically distinct tetramers. Within each tetramer there are two general types of molecule, one (**A**, **C**, and **F**) which is hydrogen-bonded to only two other molecules, and which for convenience is referred to as terminal, and the other (**B**, **D**, and **E**) which is hydrogen-bonded to three other molecules and is referred to as bridging. Within a tetramer the bridging molecules are hydrogen-bonded to form dimeric units, similar to those found in $R_2Si(OH)_2$ with R = i-Pr, C_6H_{11}, or t-Bu, shown diagrammatically in I. The dimeric units are not, however, linked together to form chains, but are hydrogen-bonded to

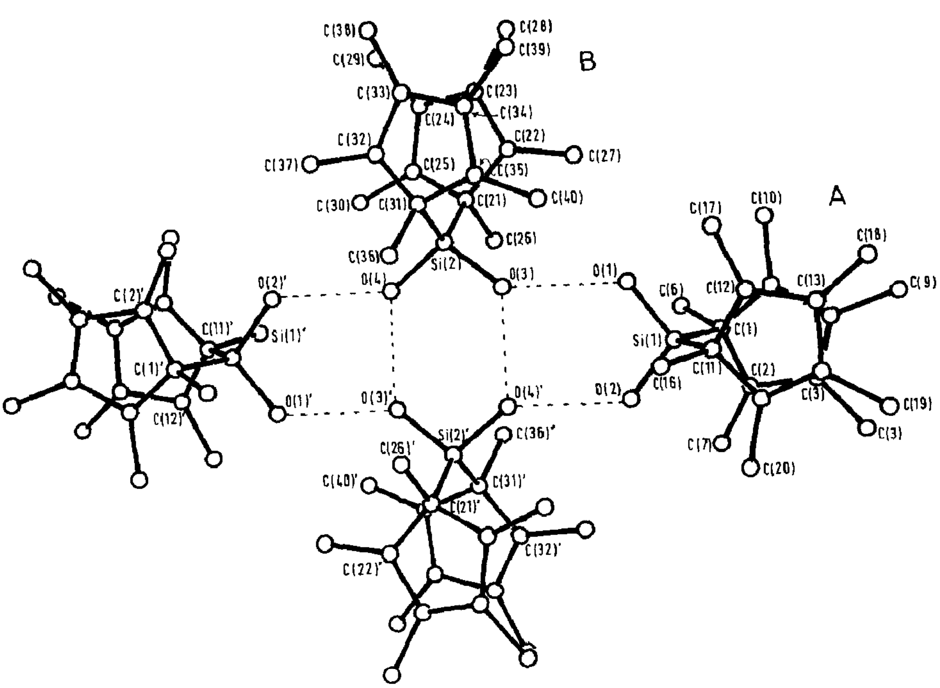

Fig. 1. An ORTEP [9] drawing of the molecular structure of and hydrogen-bonding in the symmetrical tetramer of $(C_5Me_5)_2Si(OH)_2$, together with the atom numbering scheme.

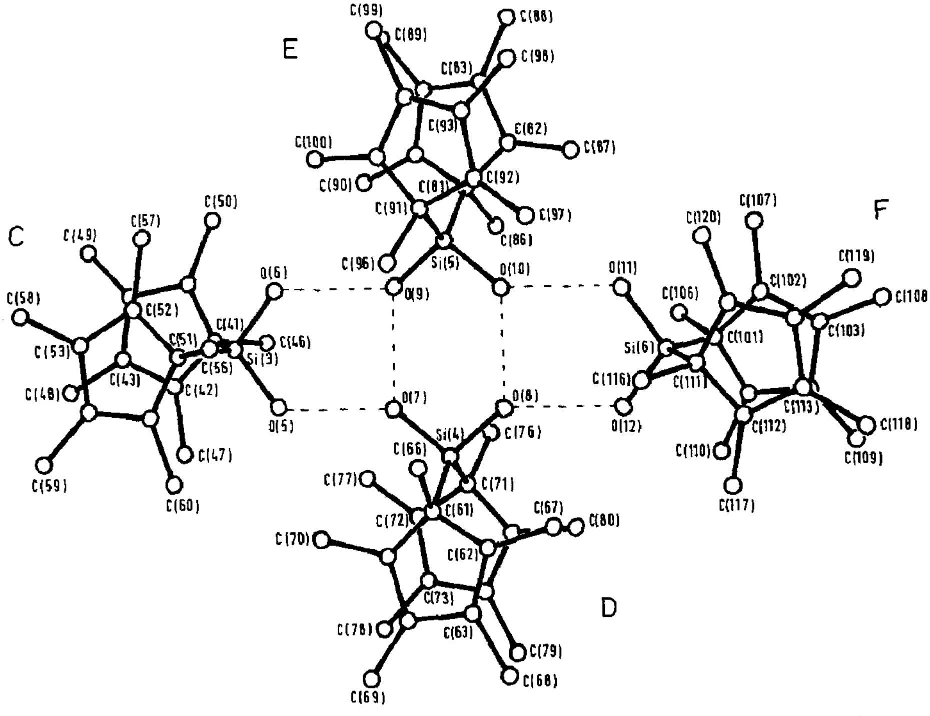

Fig. 2. An ORTEP [9] drawing of the molecular structure of and hydrogen-bonding in the unsymmetrical tetramer of $(C_5Me_5)_2Si(OH)_2$, together with the atom numbering scheme.

Table 2

Selected intramolecular distances (Å) and angles (°)[a] with estimated Standard Deviations in parentheses.

Bond lengths			
Si(1)–O(1)	1.65(2)	Si(1)–O(2)	1.64(2)
Si(1)–C(1)	1.91(3)	Si(1)–C(11)	1.90(3)
Si(2)–O(3)	1.68(2)	Si(2)–O(4)	1.66(2)
Si(2)–C(21)	1.85(3)	Si(2)–C(31)	1.99(3)
Si(3)–O(5)	1.65(2)	Si(3)–O(6)	1.64(2)
Si(3)–C(41)	1.84(3)	Si(3)–C(51)	1.90(3)
Si(4)–O(7)	1.67(2)	Si(4)–O(8)	1.69(2)
Si(4)–C(61)	1.97(3)	Si(4)–C(71)	1.92(3)
Si(5)–O(9)	1.66(2)	Si(5)–O(10)	1.69(2)
Si(5)–C(81)	1.94(2)	Si(5)–C(91)	1.90(3)
Si(6)–O(11)	1.67(2)	Si(6)–O(12)	1.66(2)
Si(6)–C(101)	1.93(2)	Si(6)–C(111)	1.89(3)
Bond angles			
O(1)–Si(1)–O(2)	106(1)	O(1)–Si(1)–C(1)	106(1)
O(1)–Si(1)–C(11)	108(1)	O(2)–Si(1)–C(1)	106(1)
O(2)–Si(1)–C(11)	108(1)	C(1)–Si(1)–C(11)	122(1)
O(3)–Si(2)–O(4)	102(1)	O(3)–Si(2)–C(21)	110(1)
O(3)–Si(2)–C(31)	104(1)	O(4)–Si(2)–C(21)	112(1)
O(4)–Si(2)–C(31)	104(1)	C(21)–Si(2)–C(31)	124(1)
O(5)–Si(3)–O(6)	105(1)	O(5)–Si(3)–C(41)	106(1)
O(5)–Si(3)–C(51)	109(1)	O(6)–Si(3)–C(41)	106(1)
O(6)–Si(3)–C(51)	107(1)	C(41)–Si(3)–C(51)	122(1)
O(7)–Si(4)–O(8)	103(1)	O(7)–Si(4)–C(61)	106(1)
O(7)–Si(4)–C(71)	108(1)	O(8)–Si(4)–C(61)	107(1)
O(8)–Si(4)–C(71)	109(1)	C(61)–Si(4)–C(71)	123(1)
O(9)–Si(5)–O(10)	102(1)	O(9)–Si(5)–C(81)	106(1)
O(9)–Si(5)–C(91)	110(1)	O(10)–Si(5)–C(81)	106(1)
O(10)–Si(5)–C(91)	108(1)	C(81)–Si(5)–C(91)	123(1)
O(11)–Si(6)–O(12)	106(1)	O(11)–Si(6)–C(101)	107.3(9)
O(11)–Si(6)–C(111)	106(1)	O(12)–Si(6)–C(101)	104(1)
O(12)–Si(6)–C(111)	108(1)	C(101)–Si(6)–C(111)	124(1)
Si(1)–C(1)–C(2)	107(2)	Si(1)–C(1)–C(5)	108(2)
Si(1)–C(1)–C(6)	111(1)	Si(1)–C(11)–C(12)	113(2)
Si(1)–C(11)–C(15)	111(2)	Si(1)–C(11)–C(16)	111(2)
Si(2)–C(21)–C(22)	112(2)	Si(2)–C(21)–C(25)	111(2)
Si(2)–C(21)–C(26)	110(2)	Si(2)–C(31)–C(32)	104(2)
Si(2)–C(31)–C(35)	107(2)	Si(2)–C(31)–C(36)	109(2)
Si(3)–C(41)–C(42)	113(2)	Si(3)–C(41)–C(45)	112(2)
Si(3)–C(41)–C(46)	112(2)	Si(3)–C(51)–C(52)	108(2)
Si(3)–C(51)–C(55)	108(2)	Si(3)–C(51)–C(56)	108(2)
Si(4)–C(61)–C(62)	109(2)	Si(4)–C(61)–C(65)	106(2)
Si(4)–C(61)–C(66)	106(2)	Si(4)–C(71)–C(72)	108(2)
Si(4)–C(71)–C(75)	108(2)	Si(4)–C(71)–C(76)	112(2)
Si(5)–C(81)–C(82)	106(2)	Si(5)–C(81)–C(85)	110(2)
Si(5)–C(81)–C(86)	110(2)	Si(5)–C(91)–C(92)	106(2)
Si(5)–C(91)–C(95)	107(2)	Si(5)–C(91)–C(96)	109(2)
Si(6)–C(101)–C(102)	103(2)	Si(6)–C(101)–C(105)	110(1)
Si(6)–C(101)–C(106)	112(1)	Si(6)–C(111)–C(112)	110(2)
Si(6)–C(111)–C(115)	107(2)	Si(6)–C(111)–C(116)	109(2)

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[a] The C–C bonds in the rings are equal to 1.50(3) Å and the ring–C to Me bonds equal 1.57(3) Å. Details of the bond angles in the Me_5C_5 groups are available from P.B.H. Symmetry element: $1-x$, $1-y$, $1-z$.

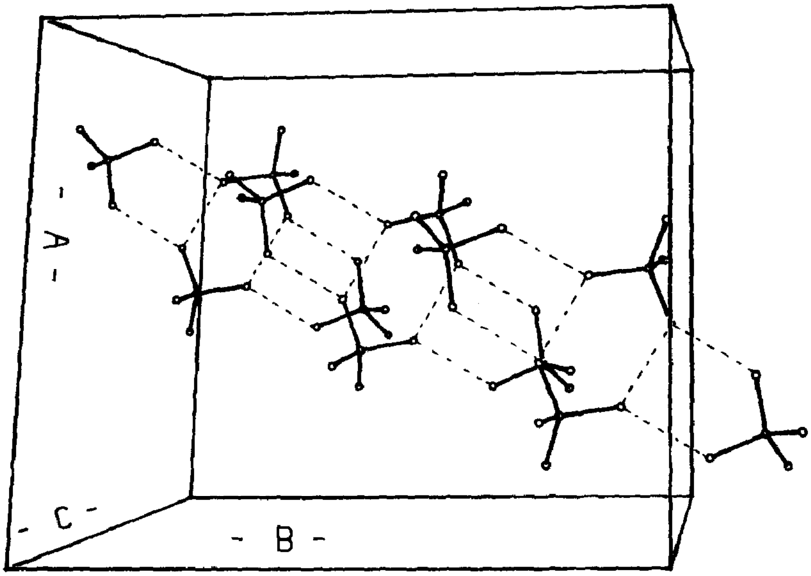

Fig. 3. Unit cell for $(C_5Me_5)_2Si(OH)_2$. Carbon atoms not bonded to silicon have been omitted for clarity.

two "chain-stopping" further molecules to form a tetramer, as shown in Figs. 1 and 2. The formation of tetramers rather than the usual ladder chains in I may be due to the steric hindrance presented by the methyl substituents on the C_5Me_5 groups, which prevent the molecules lining up alongside one another to form such chains. The tetramers could in principle be further linked through the OH groups on the terminal molecules, but this is probably prevented by the α-methyl groups, which occupy space above and below the plane of each tetramer.

There are no unusual features in the bond lengths and angles shown in Table 2. The oxygen–oxygen distance within each bridging pair, O(3)–O(4′) (2.71), O(7)–O(9) (2.73), and O(8)–O(10) (2.65 Å) are very similar to the corresponding distances in $i\text{-}Pr_2Si(OH)_2$ (2.71) [3], $t\text{-}Bu_2Si(OH)_2$ (2.69) [4], and $(C_6H_{11})_2Si(OH)_2$ (2.73 Å) [5]. The oxygen–oxygen distances between the bridging and the terminal molecules O(1)–O(3) (2.74Å) and O(2)–O(4′), O(5)–O(7), O(6)–O(9), O(8)–O(12), and O(10)–O(11) (average 2.75 Å) are slightly longer than those within the bridging pairs and are again very similar to those in $i\text{-}Pr_2Si(OH)_2$(2.74) [3], $t\text{-}Bu_2Si(OH)_2$ (2.77) [4], and $(C_6H_{11})_2Si(OH)_2$ (2.75 Å) [5]. The O–Si–O angles in the terminal molecules (*A*, *C*, and *F*) average about 106° and in the bridging molecules (*B*, *D*, and *E*) the average is about 102°, while the C–Si–C angles are all very similar, averaging 123°. The average of the O–Si–O angles is slightly smaller, and that of the C–Si–C angles is slightly larger, than those found in other silanediols.

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References

1 P.E. Tomlins, J.E. Lydon, D. Akrigg, and B. Sheldrick, Acta Crystallogr., C, 41 (1985) 941.

2 J.K. Fawcett, N. Camerman, and A. Camerman, Canad. J. Chem., 55 (1977) 3631; L. Parkanyi and B. Bocelli, Cryst. Struct. Commun., 7 (1978) 335.

3 N.H. Buttrus, C. Eaborn, P.B. Hitchcock, and P.D. Lickiss, J. Organomet. Chem., 302 (1986) 159.

4 N.H. Buttrus, C. Eaborn, P.B. Hitchcock, and A.K. Saxena, J. Organomet. Chem., 284 (1985) 291.

5 N.H. Buttrus, C. Eaborn, P.B. Hitchcock, P.D. Lickiss, and A.D. Taylor, J. Organomet. Chem., 309 (1986) 25.

6 S.S. Al-Juaid, C. Eaborn, P.B. Hitchcock, and P.D. Lickiss, J. Organomet. Chem., 362 (1989) 17.

7 Z.H. Aiube, N.H. Buttrus, C. Eaborn, P.B. Hitchcock, and J.A. Zora, J. Organomet. Chem., 292 (1985) 177.

8 P. Jutzi, D. Kanne, and C. Krüger, Angew. Chem., Int. Ed. Engl., 25 (1986) 164.

9 C.K. Johnson, Report ORNL-3794, (1965), Oak Ridge National Laboratory, Oak Ridge, Tennessee.