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The crystal structure of tetrakis(hydroxydimethylsilyl)methane. A remarkable three-dimensional hydrogen-bonded network

Salih S. Al-Juaid, Colin Eaborn, Peter B. Hitchcock, and Paul D. Lickiss *

School of Chemistry and Molecular Sciences, University of Sussex, Brighton BN1 9QJ (Great Britain)

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Abstract

The crystal structure of $C(SiMe_2OH)_4$ consists of an infinite three-dimensional array of hydrogen-bonded molecules. Each molecule contains two intramolecular hydrogen bonds, and the molecules are connected by intermolecular hydrogen bonds to form chains, which are cross linked by hydrogen bonding to form sheets, and these in turn are hydrogen-bonded together to form a three-dimensional network.

Introduction

We have recently determined the crystal structures of several organosilanediols [1–4] and observed in them several types of intermolecularly hydrogen-bonded frameworks; in none of the cases we examined were there infinite two-dimensional hydrogen-bonded layers of the type observed for $Et_2Si(OH)_2$ [5]. In the special case of the dihydroxide $(Me_3Si)_2C(SiMe_2OH)_2$ [4] each molecule contains one intramolecular hydrogen bond, and hydrogen bonding between molecules leads to infinite chains. We thus thought it of interest to determine the structure of the tetrahydroxide $C(SiMe_2OH)_4$, to see whether each molecule would, as expected by simple analogy, contain two intramolecular hydrogen bonds, and to find out how the molecules are linked by intermolecular hydrogen bonding. The results confirmed the presence of the intramolecular bonding and revealed a remarkable three-dimensional hydrogen-bonded network.

Experimental

The tetrahydroxide was prepared by treating $C(SiMe_2I)_4$ [6] with AgOCN in moist ether as described elsewhere [7]. Crystals suitable for the X-ray study were obtained by slow evaporation of diethyl ether from a heptane/ether solution.

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Table 1

Fractional atomic coordinates ($\times 10^4$) with estimated standard deviations in parentheses

Atom	x	y	z
Si(1)	6245(1)	771(1)	2829(1)
Si(2)	4586(1)	1635(1)	2523(1)
Si(3)	5451(1)	521(1)	1804(1)
Si(4)	4690(1)	−306(1)	2697(1)
Si(5)	7870(1)	2244(1)	4743(1)
Si(6)	8130(1)	4188(1)	4858(1)
Si(7)	6393(1)	3379(2)	4865(1)
Si(8)	7665(2)	3128(2)	5718(1)
Si(9)	7766(1)	−2119(1)	4334(1)
Si(10)	7807(1)	−761(1)	5098(1)
Si(11)	7567(1)	−2634(1)	5399(1)
Si(12)	6165(1)	−1714(1)	4855(1)
Si(13)	10481(2)	507(1)	6784(1)
Si(14)	9468(2)	1491(1)	7486(1)
Si(15)	11062(2)	578(1)	7862(1)
Si(16)	9560(2)	−473(1)	7559(1)
O(1)	6863(3)	−3(3)	2676(2)
O(2)	4934(3)	2336(3)	2131(2)
O(3)	6043(4)	1319(3)	1675(2)
O(4)	5456(3)	−979(3)	2803(2)
O(5)	7080(3)	1600(3)	4800(2)
O(6)	7995(4)	4883(3)	5288(2)
O(7)	5862(3)	2664(4)	5161(3)
O(8)	7162(4)	3906(4)	5971(2)
O(9)	7814(3)	−1228(3)	4032(2)
O(10)	7384(3)	−67(3)	4714(2)
O(11)	7343(4)	−3545(3)	5137(2)
O(12)	5844(3)	−2691(3)	4882(2)
O(13)	11175(4)	1258(3)	6735(2)
O(14)	9937(3)	2188(3)	7155(2)
O(15)	11663(4)	−216(3)	7737(2)
O(16)	10289(3)	−1193(3)	7675(2)
C(1)	5220(4)	657(4)	2465(3)
C(2)	6150(6)	681(6)	3493(3)
C(3)	6845(5)	1731(6)	2711(4)
C(4)	4662(6)	2138(5)	3124(3)
C(5)	3451(5)	1496(6)	2382(4)
C(6)	4518(6)	599(6)	1384(3)
C(7)	5998(6)	−448(5)	1644(3)
C(8)	3914(5)	−793(5)	2258(4)
C(9)	4119(6)	−142(6)	3267(3)
C(10)	7539(4)	3233(4)	5037(3)
C(11)	8817(5)	1740(5)	5021(4)
C(12)	8070(7)	2356(6)	4090(4)
C(13)	9261(6)	4003(6)	4810(4)
C(14)	7770(6)	4673(6)	4272(3)
C(15)	6103(7)	3233(7)	4229(4)
C(16)	5944(5)	4389(6)	5043(4)
C(17)	8756(7)	3217(6)	5954(4)
C(18)	7213(7)	2175(6)	5980(3)
C(19)	7338(4)	−1802(4)	4935(3)
C(20)	8823(5)	−2598(6)	4367(3)
C(21)	7123(6)	−2865(6)	3973(3)
C(22)	8957(5)	−708(5)	5052(4)

Table 1 (*Continued*)

Atom	*x*	*y*	*z*
C(23)	7581(6)	−373(6)	5719(3)
C(24)	6943(6)	−2568(6)	5949(3)
C(25)	8670(6)	−2696(6)	5625(3)
C(26)	5766(5)	−1237(6)	4278(4)
C(27)	5648(5)	−1099(6)	5325(3)
C(28)	10120(4)	528(4)	7430(3)
C(29)	11039(6)	−448(5)	6600(3)
C(30)	9649(6)	708(6)	6317(3)
C(31)	8365(6)	1374(6)	7255(5)
C(32)	9406(7)	1913(6)	8098(4)
C(33)	11678(6)	1553(5)	7839(4)
C(34)	10796(7)	390(6)	8503(3)
C(35)	8878(6)	−435(7)	8083(4)
C(36)	8894(6)	−910(6)	7050(4)

Crystal data

$C_9H_{28}O_4Si_4$, M 312.7, monoclinic, a 16.051(1), b 16.095(3), c 27.629 Å, β 92.35(1)°, U 7127 Å^3, Z 16, D_c 1.17 g cm^{-3}, $F(000)$ 2720. Monochromated Mo-K_α radiation, λ 0.71069 Å, μ 3.3 cm^{-1}. Space group $P2_1/c$ from systematic absences of $h0l$ for l odd and $0k0$ for k odd.

A crystal of ca. $0.3 \times 0.3 \times 0.25$ mm was used for data collection on an Enraf–Nonius CAD4 diffractometer. Intensities for $hk \pm l$ reflections with $2 < \theta < 20°$ were measured by a $\theta/2\theta$ scan with a scan width of $\Delta\theta = (0.8 + 0.35 \tan\theta)°$. A rapid pre-scan at 10^3 min^{-1} in θ was used to determine the scan rate for each reflection, where the reflections with $I/\sigma(I)$ less than 1 were coded as unobserved. The remainder were rescanned subject to a maximum $I/\sigma(I)$ of 20 or a maximum recording time of 60 s. Two standard reflections monitored every 30 min showed no

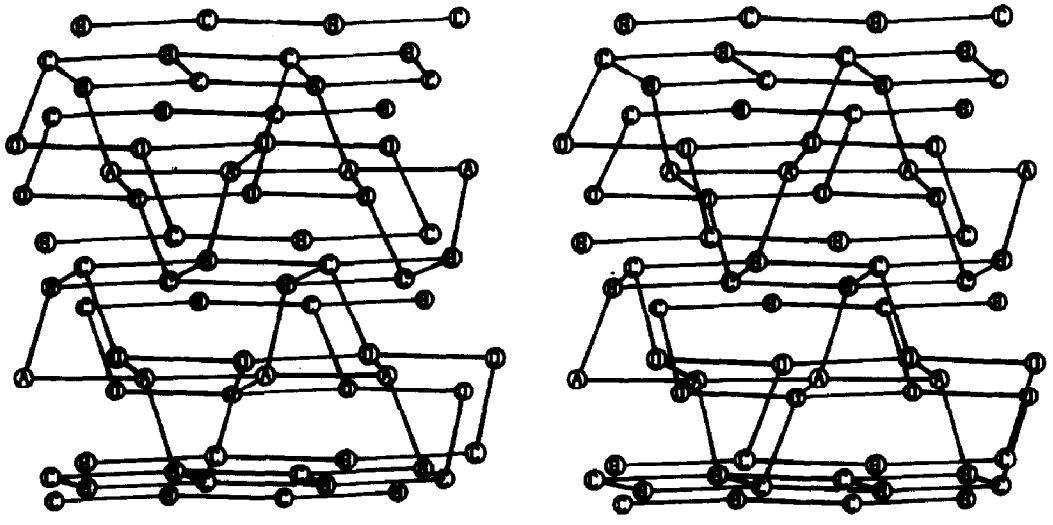

Fig. 1. Stereopair showing the pattern of intermolecular hydrogen bonding in $(HOMe_2Si)_4C$. The independent molecules **A**, **B**, **C**, and **D** are represented by circles.

Table 2

Intramolecular distances (Å) and angles (°) with estimated standard deviations in parentheses

Si(1)–O(1)	1.658(6)	Si(1)–C(1)	1.902(7)
Si(1)–C(2)	1.851(10)	Si(1)–C(3)	1.856(9)
Si(2)–O(2)	1.674(6)	Si(2)–C(1)	1.885(7)
Si(2)–C(4)	1.848(9)	Si(2)–C(5)	1.861(9)
Si(3)–O(3)	1.644(6)	Si(3)–C(1)	1.892(8)
Si(3)–C(6)	1.860(10)	Si(3)–C(7)	1.851(9)
Si(4)–O(4)	1.655(6)	Si(4)–C(1)	1.893(7)
Si(4)–C(8)	1.874(9)	Si(4)–C(9)	1.873(10)
Si(5)–O(5)	1.651(5)	Si(5)–C(10)	1.875(7)
Si(5)–C(11)	1.860(9)	Si(5)–C(12)	1.855(11)
Si(6)–O(6)	1.652(6)	Si(6)–C(10)	1.882(7)
Si(6)–C(13)	1.849(10)	Si(6)–C(14)	1.867(9)
Si(7)–O(7)	1.667(7)	Si(7)–C(10)	1.895(7)
Si(7)–C(15)	1.816(12)	Si(7)–C(16)	1.852(10)
Si(8)–O(8)	1.659(6)	Si(8)–C(10)	1.893(8)
Si(8)–C(17)	1.850(11)	Si(8)–C(18)	1.856(10)
Si(9)–O(9)	1.662(6)	Si(9)–C(19)	1.892(7)
Si(9)–C(20)	1.863(9)	Si(9)–C(21)	1.848(10)
Si(10)–O(10)	1.665(6)	Si(10)–C(19)	1.883(7)
Si(10)–C(22)	1.858(9)	Si(10)–C(23)	1.874(9)
Si(11)–O(11)	1.668(6)	Si(11)–C(19)	1.880(7)
Si(11)–C(24)	1.856(10)	Si(11)–C(25)	1.857(9)
Si(12)–O(12)	1.657(6)	Si(12)–C(19)	1.892(7)
Si(12)–C(26)	1.858(10)	Si(12)–C(27)	1.857(10)
Si(13)–O(13)	1.653(6)	Si(13)–C(28)	1.901(8)
Si(13)–C(29)	1.860(9)	Si(13)–C(30)	1.845(10)
Si(14)–O(14)	1.649(6)	Si(14)–C(28)	1.880(7)
Si(14)–C(31)	1.867(10)	Si(14)–C(32)	1.828(10)
Si(15)–O(15)	1.645(6)	Si(15)–C(28)	1.890(7)
Si(15)–C(33)	1.857(9)	Si(15)–C(34)	1.861(10)
Si(16)–O(16)	1.669(6)	Si(16)–C(28)	1.884(7)
Si(16)–C(35)	1.851(10)	Si(16)–C(36)	1.868(10)
O(1)–Si(1)–C(1)	107.8(3)	O(1)–Si(1)–C(2)	105.5(4)
O(1)–Si(1)–C(3)	105.1(4)	C(1)–Si(1)–C(2)	114.3(4)
C(1)–Si(1)–C(3)	115.5(4)	C(2)–Si(1)–C(3)	107.7(4)
O(2)–Si(2)–C(1)	108.2(3)	O(2)–Si(2)–C(4)	105.9(3)
O(2)–Si(2)–C(5)	107.2(4)	C(1)–Si(2)–C(4)	115.2(4)
C(1)–Si(2)–C(5)	114.2(4)	C(4)–Si(2)–C(5)	105.6(4)
O(3)–Si(3)–C(1)	104.8(3)	O(3)–Si(3)–C(6)	105.7(4)
O(3)–Si(3)–C(7)	108.8(4)	C(1)–Si(3)–C(6)	114.2(4)
C(1)–Si(3)–C(7)	116.1(4)	C(6)–Si(3)–C(7)	106.6(4)
O(4)–Si(4)–C(1)	104.7(3)	O(4)–Si(4)–C(8)	108.1(3)
O(4)–Si(4)–C(9)	109.2(4)	C(1)–Si(4)–C(8)	114.7(4)
C(1)–Si(4)–C(9)	114.3(4)	C(8)–Si(4)–C(9)	105.6(4)
O(5)–Si(5)–C(10)	105.1(3)	O(5)–Si(5)–C(11)	107.7(3)
O(5)–Si(5)–C(12)	108.6(4)	C(10)–Si(5)–C(11)	115.6(4)
C(10)–Si(5)–C(12)	113.8(4)	C(11)–Si(5)–C(12)	105.8(5)
O(6)–Si(6)–C(10)	106.3(3)	O(6)–Si(6)–C(13)	108.7(4)
O(6)–Si(6)–C(14)	107.2(4)	C(10)–Si(6)–C(13)	113.2(4)
C(10)–Si(6)–C(14)	115.2(4)	C(13)–Si(6)–C(14)	105.9(5)
O(7)–Si(7)–C(10)	107.6(3)	O(7)–Si(7)–C(15)	105.6(4)
O(7)–Si(7)–C(16)	105.3(4)	C(10)–Si(7)–C(15)	115.8(4)
C(10)–Si(7)–C(16)	115.3(4)	C(15)–Si(7)–C(16)	106.4(5)
O(8)–Si(8)–C(10)	108.5(3)	O(8)–Si(8)–C(17)	105.2(4)

Table 2 (*Continued*)

O(8)–Si(8)–C(18)	104.8(4)	C(10)–Si(8)–C(17)	113.8(4)
C(10)–Si(8)–C(18)	115.7(4)	C(17)–Si(8)–C(18)	107.9(5)
O(9)–Si(9)–C(19)	103.5(3)	O(9)–Si(9)–C(20)	108.7(4)
O(9)–Si(9)–C(21)	109.0(4)	C(19)–Si(9)–C(20)	115.6(4)
C(19)–Si(9)–C(21)	115.7(4)	C(20)–Si(9)–C(21)	104.1(4)
O(10)–Si(10)–C(19)	107.2(3)	O(10)–Si(10)–C(22)	107.8(4)
O(10)–Si(10)–C(23)	105.6(4)	C(19)–Si(10)–C(22)	114.4(4)
C(19)–Si(10)–C(23)	115.1(4)	C(22)–Si(10)–C(23)	106.1(4)
O(11)–Si(11)–C(19)	107.3(3)	O(11)–Si(11)–C(24)	107.0(4)
O(11)–Si(11)–C(25)	106.5(4)	C(19)–Si(11)–C(24)	114.8(4)
C(19)–Si(11)–C(25)	115.1(4)	C(24)–Si(11)–C(25)	105.5(4)
O(12)–Si(12)–C(19)	103.5(3)	O(12)–Si(12)–C(26)	109.6(4)
O(12)–Si(12)–C(27)	109.0(4)	C(19)–Si(12)–C(26)	116.0(4)
C(19)–Si(12)–C(27)	115.3(4)	C(26)–Si(12)–C(27)	103.4(4)
O(13)–Si(13)–C(28)	107.3(3)	O(13)–Si(13)–C(29)	104.4(4)
O(13)–Si(13)–C(30)	106.6(4)	C(28)–Si(13)–C(29)	116.0(4)
C(28)–Si(13)–C(30)	114.4(4)	C(29)–Si(13)–C(30)	107.3(4)
O(14)–Si(14)–C(28)	104.4(3)	O(14)–Si(14)–C(31)	109.1(4)
O(14)–Si(14)–C(32)	107.7(4)	C(28)–Si(14)–C(31)	114.3(4)
C(28)–Si(14)–C(32)	115.7(4)	C(31)–Si(14)–C(32)	105.4(5)
O(15)–Si(15)–C(28)	107.2(3)	O(15)–Si(15)–C(33)	109.4(4)
O(15)–Si(15)–C(34)	103.5(4)	C(28)–Si(15)–C(33)	115.3(4)
C(28)–Si(15)–C(34)	112.6(4)	C(33)–Si(15)–C(34)	108.3(5)
O(16)–Si(16)–C(28)	107.1(3)	O(16)–Si(16)–C(35)	107.7(4)
O(16)–Si(16)–C(36)	105.0(4)	C(28)–Si(16)–C(35)	114.9(4)
C(28)–Si(16)–C(36)	116.3(4)	C(35)–Si(16)–C(36)	105.2(5)
Si(1)–C(1)–Si(2)	109.4(4)	Si(1)–C(1)–Si(3)	108.8(4)
Si(1)–C(1)–Si(4)	106.9(4)	Si(2)–C(1)–Si(3)	107.8(4)
Si(2)–C(1)–Si(4)	113.8(4)	Si(3)–C(1)–Si(4)	110.0(4)
Si(5)–C(10)–Si(6)	115.2(4)	Si(5)–C(10)–Si(7)	106.6(4)
Si(5)–C(10)–Si(8)	109.5(4)	Si(6)–C(10)–Si(7)	109.1(4)
Si(6)–C(10)–Si(8)	107.4(4)	Si(7)–C(10)–Si(8)	108.8(4)
Si(9)–C(19)–Si(10)	107.1(4)	Si(9)–C(19)–Si(11)	109.7(4)
Si(9)–C(19)–Si(12)	108.2(4)	Si(10)–C(19)–Si(11)	113.9(4)
Si(10)–C(19)–Si(12)	110.4(4)	Si(11)–C(19)–Si(12)	107.4(4)
Si(13)–C(28)–Si(14)	106.4(4)	Si(13)–C(28)–Si(15)	109.1(4)
Si(13)–C(28)–Si(16)	109.2(4)	Si(14)–C(28)–Si(15)	110.3(4)
Si(14)–C(28)–Si(16)	114.7(4)	Si(15)–C(28)–Si(16)	107.0(4)

significant variation. After correction for Lorentz and polarization effects but not for absorption, equivalent data were averaged and 4012 reflections with $|F^2| > \sigma(F^2)$ were used for the structure analysis. The values of $\sigma(F^2)$ were calculated as $[\sigma^2(I) + (0.04I)^2]^{1/2}/Lp$.

There were found to be four independent molecules in the asymmetric unit of the crystal. The positions of C, Si, and O atoms were found by direct methods, and refined by full matrix least squares with anisotropic temperature factors. The hydrogen atoms of the Me groups were placed at calculated positions (C–H 1.08 Å) and held fixed with $B_{iso} = 6.0$ Å^2. The hydrogen atoms of the hydroxyl groups could not be located and were not included. Further refinement converged at $R = 0.056$, $R' = 0.061$ with weighting scheme $w = 1/\sigma^2(F)$, and a final shift to error ratio < 0.14. A final difference map had peaks of up to 0.3 eÅ^{-3}.

The structure solution and refinement were carried out on a PDP 11/34 computer using the Enraf–Nonius Structure Determination Package. Scattering factors for neutral atoms were taken from ref. 8. Final atom coordinates are listed in Table 1 and bond lengths and angles in Table 2. A table of temperature factors and lists of structure factors are available from the authors.

Results and discussion

In discussing the hydrogen bonding it is convenient to refer to the four independent molecules centred on atoms C(1), C(10), C(19), and C(28) as molecules **A**, **B**, **C**, and **D** respectively. The unit cell can be regarded as having layers of molecules parallel to the (001) plane, with the layers at $z = 0$ and $z = 1/2$ containing only molecules **B** and **C**, and the layers at $z = 1/4$ and $z = 3/4$ containing molecules **A** and **D**. Within each molecule there are two pairs of intramolecularly hydrogen-

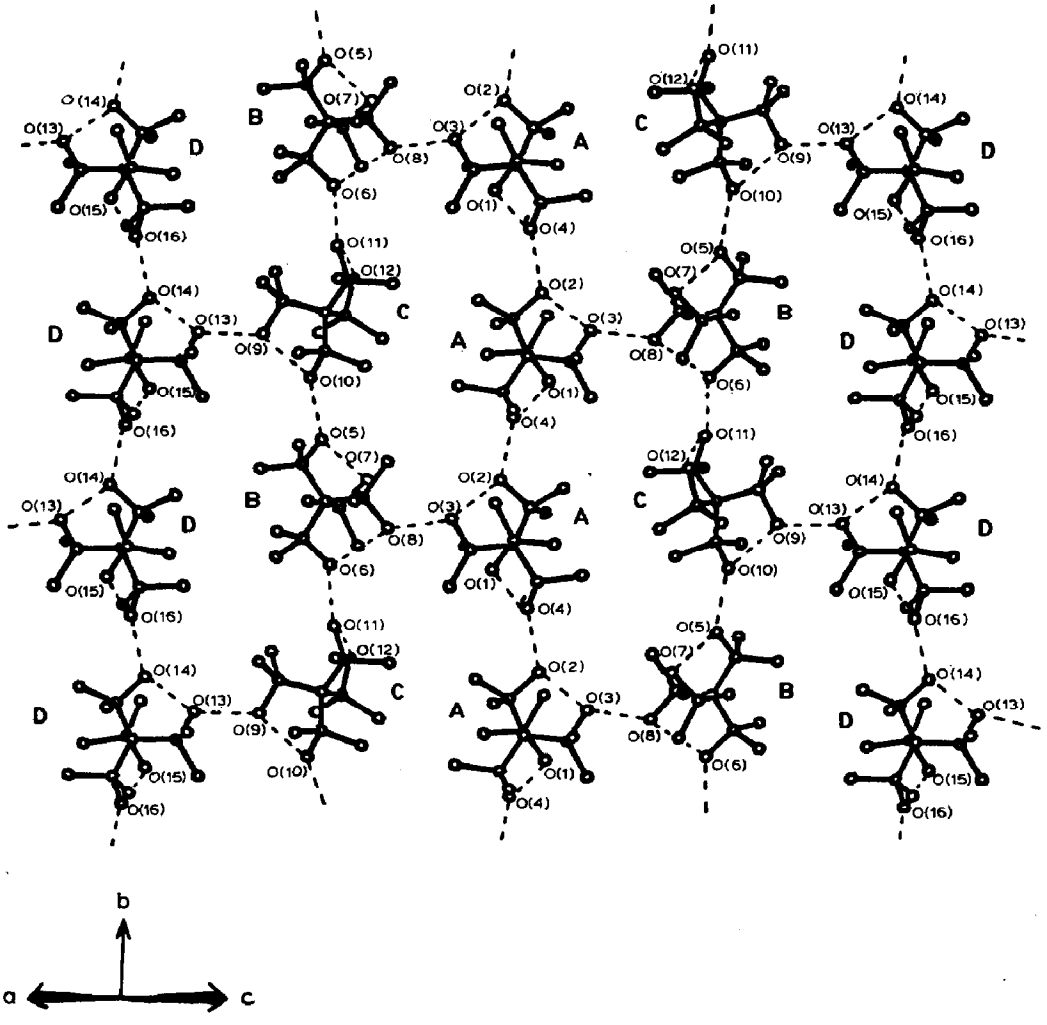

Fig. 2. Hydrogen bonding between molecules of type **A**, **B**, **C**, and **D** in the (101) plane.

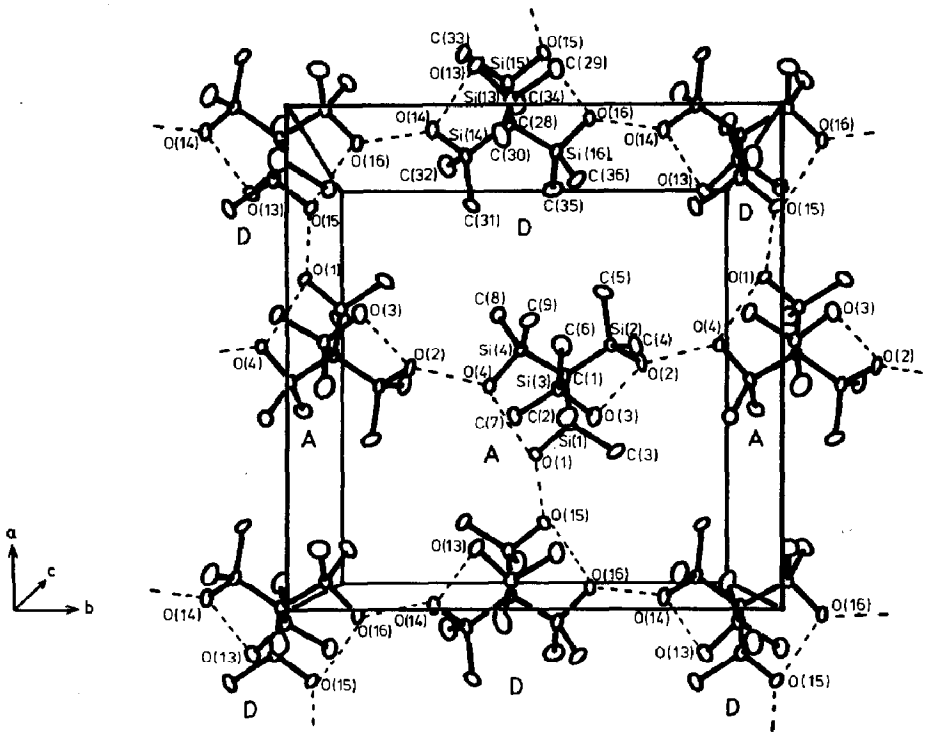

Fig. 3. Hydrogen bonding between molecules of type **A** and **D** in the (001) plane.

bonded oxygen atoms; one of these pairs lies roughly in the (001) plane and the other in the (101) plane. Each oxygen then also makes an intermolecular hydrogen bond to another molecule. The pattern of intermolecular hydrogen bonding is shown in the stereopair (Fig. 1), in which the molecules are represented only by a single point and only the intermolecular hydrogen bonds are shown. It can be seen that there are sheets of hydrogen bonds in a "chicken-wire" arrangement parallel to the (101) planes, with cross-linking hydrogen bonds between these sheets in the (001) plane at $z = 0$, $1/4$, $1/2$, and $3/4$. These three planes are shown separately in Figs. 2, 3 and 4.

Figure 2 shows the hydrogen bonds joining the molecules of type **A**, **B**, **C** and **D** in the plane (101). Each molecule has two intramolecular hydrogen bonds similar to those in the diol $(Me_3Si)_2C(SiMe_2OH)_2$, one being in the (101) plane (with both oxygen atoms also involved in intermolecular hydrogen bonds within the plane) and the other is at an angle to the plane, and has the oxygen at one end involved in an intermolecular hydrogen bond in the plane and the other (O(1), O(7), O(12), or O(15)) involved in an intermolecular hydrogen bond to the adjacent sheets. In the figure, on the left, there is a chain of **D** molecules, parallel to the *b* axis, with the out of plane hydrogen bonds from O(15) alternately above and below the plane. Next is a chain of alternating **B** and **C** molecules with the out of plane hydrogen bonds from O(7) and O(12) all below the plane. Then there is a chain of **A** molecules with out of plane hydrogen bonds from O(1) alternately above and below the plane. Next is a chain of alternating **B** and **C** molecules with out of plane hydrogen bonds from O(7) and O(12) all above the plane. Finally there is a repeat chain of **D** molecules. The adjacent sheets above and below this plane will have a chain of **D** above and

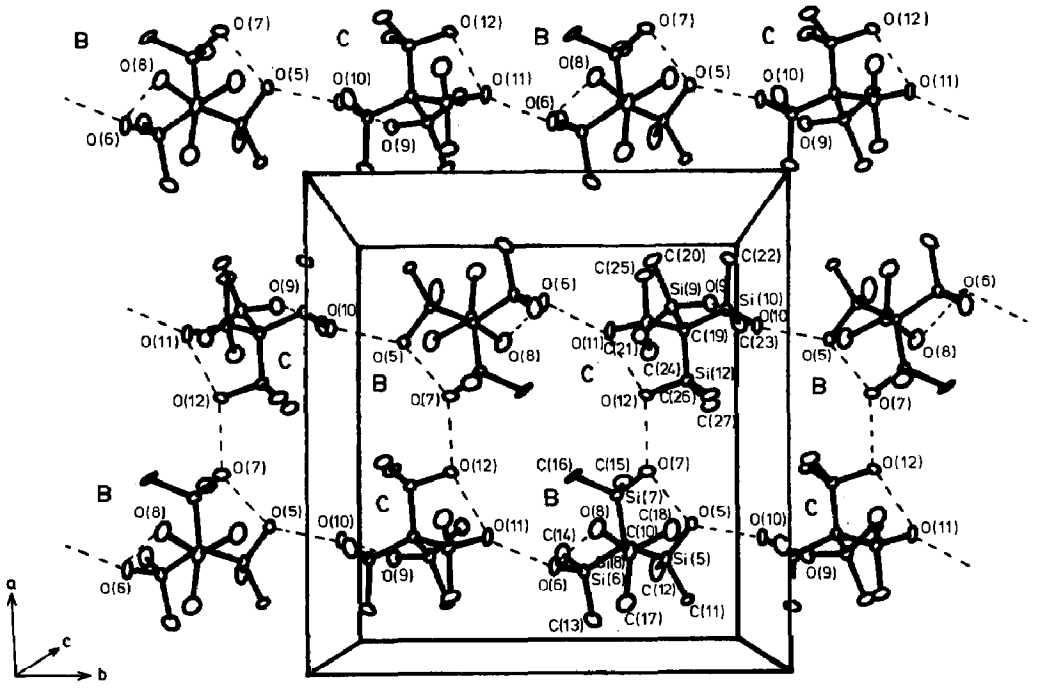

Fig. 4. Hydrogen bonding between molecules of type **B** and **C** in the (001) plane.

below the chain of **A** molecules in the reference plane, a chain of **A** above and below the chain of **D**, and **BC** chains above and below the **BC** chains in the reference plane.

Figure 3 shows the hydrogen bonds joining the **A** and **D** molecules in the (001) plane at $z = 1/4$. The pattern of hydrogen bonding is similar to that in the (101) plane. The plane at $z = 3/4$ is similar.

Figure 4 shows the hydrogen bonds joining the **B** and **C** molecules in the (001) plane at $z = 1/2$. The pattern of hydrogen-bonds is different in this case, with alternate pairs of chains parallel to b either cross linked at every every molecule or not at all. The plane at $z = 0$ is similar.

The silicon to central carbon bond lengths are normal, and average 1.893(6), 1.886(8), 1.886(5), and 1.888(8) Å for molecules **A**, **B**, **C**, and **D**, respectively. The six Si–C–Si bond angles around the central carbon atom in each distinct molecule fall into three types: (i) two angles smaller than 109° (averaging 107.4(4), 107.0(4), 107.3(4), and 106.7(4)° in molecules **A**, **B**, **C** and **D**, respectively); (ii) three angles of approximately 109° (averaging 109.4(5), 109.1(3), 109.4(8), and 109.5(5)° in molecules **A**, **B**, **C**, and **D**, respectively); and (iii) one angle larger than 109° (113.8(4), 115.2(4), 113.9(4), and 114.7(4)° in molecules **A**, **B**, **C**, and **D**, respectively). The three types correspond to: (1) the two pairs of silicons on which OH groups are hydrogen-bonded intramolecularly to each other e.g. Si(2)–C(1)–Si(3) in molecule **A**, with the SiCSi angles compressed below 109°; (2) the three SiCSi angles which do not form part of the hydrogen-bonded chain of molecules, e.g. Si(2)–C(1)–Si(1); and (3) the SiCSi angle between the pair of silicons within a molecule that are involved in the intermolecular chain-forming hydrogen bonding, e.g. Si(2)–C(1)–Si(4) in molecule **A**.

It is noteworthy that pentaerythritol, $C(CH_2OH)_4$, which has the potential for forming a similar network, has a very different structure. It contains no intramolecular hydrogen bonds, and strong intermolecular hydrogen bonds join the molecules to form planes, with only weak interactions between the planes [9,10].

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