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The Structures of 1-Deoxy-(*N*-methyloctanamido)-D-glucitol (MEGA-8) and 1-Deoxy-(*N*-methylundecanamido)-D-glucitol (MEGA-11)

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Abstract

1-Deoxy-(*N*-methyloctanamido)-D-glucitol, C₁₅H₃₁NO₆ (MEGA-8), crystallizes in space group *P*2₁2₁2₁, *M*_r = 321.4, *a* = 4.865 (1), *b* = 9.186 (3), *c* = 39.097 (9) Å, *V* = 1747.24 Å³, *Z* = 4, *D*_x = 1.22 Mg m⁻³, λ(Cu *K*α) = 1.5418 Å, μ = 0.78 mm⁻¹, *F*(000) = 704, *R* = 0.035 for 1318 reflections. 1-Deoxy-(*N*-methylundecanamido)-D-glucitol, C₁₈H₃₇NO₆ (MEGA-11), crystallizes in space group *P*1, *M*_r = 363.5, *a* = 4.950 (1), *b* = 5.6027 (8), *c* = 19.162 (4) Å, α = 83.19 (2), β = 89.76 (2), γ = 76.28 (2)°, *V* = 512.64 Å³, *Z* = 1, *D*_x = 1.18 Mg m⁻³, λ(Mo *K*α) = 0.7107 Å, μ = 0.093 mm⁻¹, *F*(000) = 200, *R* = 0.061 for 1898 reflections. The glucitol C-atom-chain conformation is different in the two structures. In MEGA-8 it is fully extended, whereas in MEGA-11 it is bent. The alkyl C-atom chains are fully extended in both structures. The molecular packing is different. In MEGA-8 it is head-to-head bilayer with intercalating alkyl chains, whereas in MEGA-11 it is monolayer head-to-tail with non-intercalating alkyl chains. The hydrogen bonding in MEGA-8 is a finite chain; in MEGA-11 it includes a homodromic four-bond cycle.

Introduction

The alkylated saccharides constitute a large class of amphiphilic molecules, many of which form, or are expected to form, both thermotropic and lyotropic liquid crystals (Jeffrey, 1986). The rod-shaped mesogens consist of a carbohydrate head group and an *n*-alkyl chain which generally has to exceed six C

atoms. It is important that the head group contains sufficient unblocked hydroxyls to form the hydrogen-bonded 'core', which is responsible for the molecular association in the liquid-crystal phase (Jeffrey, 1984; Goodby, 1984). In the carbohydrates, the head group may be cyclic, *i.e.* a pyranosyl or furanosyl ring, bicyclic, as in a disaccharide, or acyclic, as in the derivatives of a sugar alcohol (Pffannemüller & Welte, 1985; Pffannemüller, Welte, Chin & Goodby, 1986; Baeyens-Volant, Cuvelier, Fornasier, Szalai & David, 1985). There are also 'disc-shaped' carbohydrate mesogens, where the central core is a pyranose or inositol ring around which there are substituted at least three and generally more alkyl chains (Kohne & Praefcke, 1984; Zimmermann, Jameson & Weiss, 1985). The functional groups, which attach the alkyl chains to the head group, appear to be irrelevant to the mesogeneity and hence great configurational variety is possible. The alkylated saccharides form lyotropic liquid crystals with water, when sufficiently soluble. Octyl β-D-glucopyranoside, a well known surfactant used in the crystallization of membrane proteins (Baron & Thompson, 1975), has classical lyotropic behavior forming laminar, cubic and hexagonal phases and a micellar solution with increasing dilution with water at room temperature (Chung & Jeffrey, 1989). The MEGA series of surfactants from MEGA-8 to MEGA-12 are also used in membrane protein crystallization (Hildreth, 1982).

This type of compound also occurs, with much more configurational complexity, in natural products, in the lipids of *Mycobacterium tuberculosis* in the form of 'cord-factors' (Asselineau & Asselineau, 1978), which

are the mycollic esters of α,α -trehalose, and in the terminal lipids of the endotoxic lipopolysaccharides obtained from salmonella *E. coli*, which are mycollic esters of an *N*-acetylglucosamine disaccharide (Labischinski, Barnickel, Bradaczek, Naumann, Rietschel & Giesbrecht, 1985).

The synthesis of these alkylated sugars has received considerable attention in recent years because of their use in the solubilization of membrane proteins (Stubbs, Smith & Litman, 1976; Lin, Riedel & Kinne, 1979; Hildreth, 1982).

X-ray crystal structure analyses have been reported for several of these compounds, *i.e.* 1-decyl α -D-glucopyranoside (Moews & Knox, 1976), 1-heptyl (*S*)- α -D-mannopyranoside (Carter, Ruble & Jeffrey, 1982), 1-octyl (*S*)- β -D-xylopyranoside (Bhattacharjee & Jeffrey, 1983), anhydrous 1-octyl α -D-glucopyranoside (van Koningsveld, Jansen & Straathof, 1988) and its mono- and hemihydrate (Jeffrey, Yeon & Abola, 1987), and 1-octyl D-gluconamide (Bhattacharjee, Jeffrey & Goodby, 1985; Zabel, Müller-Fahrnow, Hilgenfeld, Saenger, Pffannemüller, Enkelmann & Welte, 1986). The series of 1-deoxy-(*N*-methylalkanamido)-D-glucitols, MEGA-8 to -12 (Hildreth, 1982) are particularly interesting from the point of view of the crystal structures, because the nonanamido derivative (MEGA-9)* has been shown to have a head-to-tail monolayer molecular packing in the crystal (Müller-Fahrnow, Zabel, Steifa & Hilgenfeld, 1986). The head-to-tail monolayer molecular packing is also observed in the crystal structures of *N*-(*n*-heptyl) and *N*-(*n*-decyl)-D-gluconamides (Müller-Fahrnow, Hilgenfeld, Hesse, Saenger & Pffannemüller, 1988). In all the alkyl pyranoside crystal structures noted above, the molecular packing was the expected head-to-head bilayer type, and related bilayer structures are inferred for the thermotropic liquid-crystal phases (Gray & Goodby, 1984).

All molecules with amphiphilic configurations do not necessarily form liquid crystals. Examples of compounds which are not mesogenic, but have crystal structures very similar to the carbohydrate mesogens are 6-*n*-hexylaminopurine and 6-*n*-heptylthiopurine (Maluszynska & Jeffrey, 1987), and octyl gallate dihydrate (Jeffrey & Yeon, 1989). An appropriate balance between the stronger stacking, dipolar or hydrophilic intermolecular forces of the core and the weaker hydrophobic binding of the alkyl tails appears to be necessary. This property is also important in the study of surfactants and has been expressed by an empirical quantity called the hydrophile-lipophile balance (HLB) (Griffin, 1949, 1954).

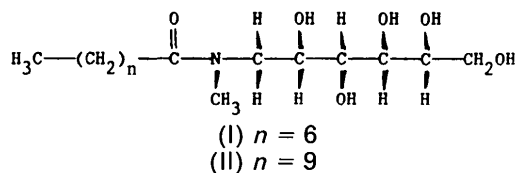
In this work, we report the crystal structures of an even and an odd member of the MEGA-*n* series:

Table 1. *Crystal and experimental data for 1-deoxy-(N-methyloctanamido)-D-glucitol (MEGA-8) and 1-deoxy-(N-methylundecanamido)-D-glucitol (MEGA-11) at 295 K*

Crystal data	MEGA-8	MEGA-11
Formula	C ₁₅ H ₃₁ NO ₆	C ₁₈ H ₃₇ NO ₆
Molecular weight	321.40	363.5
Space group	P2 ₁ 2 ₁ 2 ₁	P1
Z	4	1
Cell dimensions		
<i>a</i> (Å)	4.865 (1)	4.950 (1)
<i>b</i> (Å)	9.186 (3)	5.6027 (8)
<i>c</i> (Å)	39.097 (9)	19.162 (4)
α (°)		83.19 (2)
β (°)		89.76 (2)
γ (°)		76.28 (2)
<i>V</i> (Å ³)	1747.24	512.64
	based on 25 reflections with $9 \leq \theta \leq 29^\circ$ using Cu K α radiation, Ni filter, $\lambda = 1.5418 \text{ \AA}$	based on 25 reflections with $8.6 \leq \theta \leq 17.8^\circ$ using Mo K α radiation, Nb filter, $\lambda = 0.7107 \text{ \AA}$
μ (cm ⁻¹)	7.8 (Cu K α)	0.93 (Mo K α)
<i>D_x</i> (g cm ⁻³)	1.22	1.18
Experimental data		
Crystal dimensions (mm)	0.06 × 0.15 × 0.23	0.07 × 0.2 × 0.5
Diffractometer	CAD-4, ω -2 θ scan	
Number of independent reflections	1582	3066
Number of observed reflections	1318, $I > 2\sigma(I)$	1899, $I > 1\sigma(I)$
Range of <i>h, k, l</i>	$0 \leq h \leq 5$ $0 \leq k \leq 10$ $0 \leq l \leq 43$	$-6 \leq h \leq 6$ $-7 \leq k \leq 0$ $-27 \leq l \leq 27$
θ_{\max} (°)	60	30
Obs./parameter ratio	5.0	7.8
Residual electron difference density (e Å ⁻³)	±0.12	±0.24
Final agreement factors		
<i>R</i> (<i>F</i>)	0.035	0.061
<i>wR</i> (<i>F</i>)	0.048	0.045
<i>S</i> (goodness of fit)	1.25	1.29

Structures determined by MITHRIL (Gilmore, 1983). Function minimized: $R = \sum [w(k|F_o| - |F_c|)^2]$, using UPALS (Lundgren, 1978). $w = 1/[\sigma^2(F) + (0.02F_o)^2]$ for MEGA-8, $w = 1/\sigma^2(F)$ for MEGA-11. $\sigma(F)$ based on counting statistics.

1-deoxy-(*N*-methyloctanamido)-D-glucitol (I) and 1-deoxy-(*N*-methylundecanamido)-D-glucitol (II). The thermotropic properties of the series MEGA-4 to MEGA-10 have been reported by Goodby, Marcus, Chin, Finn & Pffannemüller (1988).



Experimental

MEGA-8 was obtained from Sigma Chemical Company and recrystallized from water. MEGA-11 was

* This compound was incorrectly named nonanoyl *N*-methyl glucamide, but it is a nonanamido derivative of D-glucitol.

Table 2. Atomic coordinates and equivalent isotropic temperature factors for MEGA-8 and MEGA-11

$$B_{eq} = \frac{4}{3} \sum_i \sum_j B_{ij}(\mathbf{a}_i, \mathbf{a}_j).$$

	x	y	z	B_{eq}/B_{iso}
MEGA-8				
C1	0.4890 (6)	0.3953 (3)	0.81417 (7)	2.94 (7)
C2	0.3104 (6)	0.3597 (3)	0.78311 (6)	2.50 (5)
O2	0.1402 (4)	0.2359 (2)	0.78920 (5)	2.91 (5)
C3	0.4944 (6)	0.3415 (3)	0.75170 (8)	2.61 (5)
O3	0.6519 (4)	0.2097 (2)	0.75415 (5)	3.30 (5)
C4	0.3380 (7)	0.3391 (3)	0.71790 (7)	2.46 (5)
O4	0.1736 (5)	0.2117 (2)	0.71365 (6)	3.14 (5)
C5	0.5297 (6)	0.3401 (3)	0.68694 (7)	2.70 (6)
O5	0.6938 (5)	0.4685 (2)	0.68870 (5)	3.18 (5)
C6	0.3774 (7)	0.3318 (3)	0.65332 (7)	3.22 (7)
O6	0.1666 (5)	0.4382 (3)	0.64970 (5)	3.65 (5)
N	0.3463 (5)	0.3854 (3)	0.84683 (5)	2.94 (5)
CN	0.1259 (8)	0.4888 (4)	0.85352 (9)	3.74 (8)
C7	0.4199 (7)	0.2820 (3)	0.86943 (7)	3.06 (7)
O7	0.6206 (5)	0.2032 (2)	0.86438 (5)	4.16 (5)
C8	0.2542 (7)	0.2687 (4)	0.90212 (8)	4.01 (8)
C9	0.3586 (10)	0.1446 (5)	0.92444 (9)	5.6 (1)
C10	0.2141 (9)	0.1340 (5)	0.9588 (1)	5.3 (1)
C11	0.3290 (11)	0.0127 (5)	0.9807 (1)	6.4 (1)
C12	0.1997 (10)	-0.0051 (5)	1.0154 (1)	6.0 (1)
C13	0.3206 (12)	-0.1288 (6)	1.0362 (1)	8.0 (2)
C14	0.1958 (17)	-0.1526 (8)	1.0704 (1)	8.6 (2)
HO2	0.240 (8)	0.163 (4)	0.7915 (9)	6 (1)
HO3	0.777 (7)	0.226 (4)	0.7677 (8)	4 (1)
HO4	0.071 (9)	0.208 (5)	0.7284 (9)	6 (1)
HO5	0.828 (8)	0.461 (4)	0.6755 (8)	5 (1)
HO6	0.230 (8)	0.525 (4)	0.6472 (9)	6 (1)
MEGA-11				
C1	-0.1534 (10)	0.1402 (9)	-0.1496 (3)	2.9 (1)
C2	-0.3777 (10)	0.0660 (8)	-0.1903 (3)	2.4 (1)
O2	-0.3881 (9)	-0.1808 (7)	-0.1646 (2)	3.4 (1)
C3	-0.3190 (10)	0.0680 (9)	-0.2674 (3)	2.4 (1)
O3	-0.5397 (9)	0.0012 (8)	-0.3034 (2)	3.3 (1)
C4	-0.2906 (10)	0.3173 (9)	-0.3045 (3)	2.3 (1)
O4	-0.5332 (9)	0.5046 (7)	-0.2936 (2)	3.1 (1)
C5	-0.2332 (10)	0.3122 (9)	-0.3818 (3)	2.5 (1)
O5	0.0022 (9)	0.1159 (7)	-0.3938 (2)	2.9 (1)
C6	-0.2009 (11)	0.5574 (9)	-0.4187 (3)	2.9 (1)
O6	0.0337 (9)	0.6166 (8)	-0.3902 (2)	3.7 (1)
N	-0.1815 (9)	0.0919 (8)	-0.0743 (2)	2.9 (1)
CN	-0.4232 (11)	0.2436 (10)	-0.0446 (3)	4.3 (1)
C7	0.0084 (10)	-0.1015 (9)	-0.0366 (3)	2.8 (1)
O7	0.2095	-0.2132	-0.0600	4.5 (1)
C8	-0.0970 (11)	-0.1815 (9)	0.0364 (3)	3.3 (1)
C9	0.0987 (11)	-0.4180 (10)	0.0703 (3)	4.3 (1)
C10	-0.0038 (11)	-0.5340 (10)	0.1374 (3)	3.9 (1)
C11	0.2050 (12)	-0.7639 (11)	0.1707 (3)	4.3 (1)
C12	0.1002 (12)	-0.9052 (11)	0.2329 (3)	4.2 (1)
C13	0.3161 (12)	-1.1316 (10)	0.2650 (3)	4.3 (1)
C14	0.2130 (11)	-1.2804 (11)	0.3261 (3)	4.3 (1)
C15	0.4275 (12)	-1.5046 (10)	0.3584 (3)	4.4 (1)
C16	0.3244 (14)	-1.6488 (12)	0.4205 (3)	5.7 (2)
C17	0.5329 (16)	-1.8845 (12)	0.4491 (4)	6.4 (2)
HO2	-0.497 (9)	-0.152 (8)	-0.133 (2)	3 (1)
HO3	-0.522 (12)	-0.153 (11)	-0.290 (3)	6 (2)
HO4	-0.640 (10)	0.512 (10)	-0.327 (3)	5 (2)
HO5	0.137 (10)	0.143 (9)	-0.369 (3)	4 (1)
HO6	-0.006 (11)	0.772 (11)	-0.393 (3)	5 (1)

obtained from OXYL, Bobingen, Federal Republic of Germany, and recrystallized from methanol. The crystal and experimental data for both compounds are given in Table 1. Both crystal structures were solved using the program *MITHRIL* (Gilmore, 1983). All the non-H atoms appeared on the *E* maps.

For MEGA-8, all initial H-atom coordinates were derived from difference maps, except those of the methylenes, which were calculated using C-H=1.0 Å and C-C-H=110°. The positional parameters for the hydroxyl and terminal methyl groups and isotropic temperature parameters for all the H atoms were

included in the full-matrix anisotropic refinement of the non-H atoms. A very strong 002 reflection for which $|\Delta F|/\sigma > 10$ was excluded from the refinement.

For MEGA-11, all H atoms were located on difference syntheses except H8, H8', H10 and H11' on the alkyl chain. The refinement parameters were the same as for MEGA-8. The very strong 001 and 007 reflections for which $|\Delta F|/\sigma > 10$ were excluded from the refinement. Atomic scattering factors were taken from *MITHRIL*. The final positional parameters for both structures are given in Table 2. The corresponding bond lengths, valence angles and torsion angles for both molecules are given in Table 3.* The atomic notation and thermal ellipsoids are shown in Fig. 1.

Discussion

The molecular and crystal structures

In both structures, the alkyl chains are fully extended with the amide group *trans*, but the D-glucitol chain conformations are different. In MEGA-8, the alditol chain is fully extended from C1 to C6. In MEGA-11, the chain has one of the two 'bent' conformations for

* Table 3 and lists of anisotropic thermal parameters, positional and temperature parameters for H atoms and observed and calculated structure factors have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 51819 (31 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

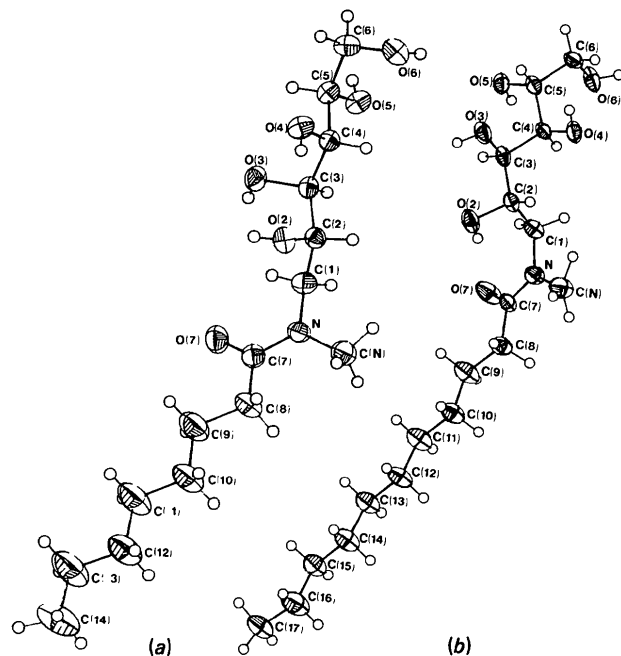
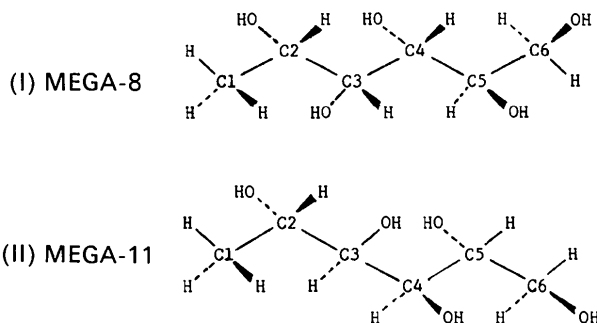


Fig. 1. Atomic notation and thermal ellipsoids (50% probability). (a) 1-Deoxy-(*n*-methyloctanamido)-D-glucitol (MEGA-8). (b) 1-Deoxy-(*N*-methylundecanamido)-D-glucitol (MEGA-11).

D-glucitol which are most stable in the absence of intramolecular hydrogen bonding (*i.e.* MAA in Jeffrey & Kim, 1970). The bend is at C2–C3 with C1–C2–C3–C4 = –58.4 (3)°.



The same straight-chain conformation is observed in the *A* form of potassium D-gluconate (Panagiotopoulos, Jeffrey, La Placa & Hamilton, 1974), where, as in this structure, it is stabilized by an O4–H...O2 intramolecular hydrogen bond. The bent-chain conformation of MEGA-11 is observed in the crystal structure of D-glucitol (Park, Jeffrey & Hamilton, 1971) and in the D-glucitol–pyridine complex (Kim, Jeffrey & Rosenstein, 1971). In both molecules, the terminal primary alcohol group is synclinal, with C4–C5–C6–O6 = +52.3 (3) and +63.7 (6)°, respectively. Since MEGA-11 is isostructural with MEGA-9 (Müller-Fahrnow *et al.*, 1986), this difference in the glucitol conformations is likely to be a characteristic difference between the members of the odd and even alkyl chain series. In the alkyl gluconamides, the straight-chain gluconamide conformation is observed in

members of both odd and even series of alkyl chains (Müller-Fahrnow *et al.*, 1988).

The glucitol and alkyl chains are not coplanar due to a twist at C2–C1–N–C7, which is +113.7 (3)° in MEGA-8 and +104.7 (5)° in MEGA-11. This is accompanied by a small bend at N–C1–C2–C3 of –167.8 (2) and –168.8 (4)°, respectively. The deviations from 180° for the alkyl-C-atom torsion angles are less than 5°, except for C7–C8–C9–C10 in MEGA-11, –169.5 (5)°.

The C–C bond lengths in both structures range from 1.504 to 1.545 Å with a mean value of 1.517 Å, except for the terminal C13–C14 in MEGA-8 which is 1.485 Å. This is probably due to thermal motion since both atoms have larger U_{iso} . The N–CH₃ bond lengths are 1.448 and 1.456 Å. The C=O and C–N bond lengths differ by 10σ and 4σ, respectively, in the two structures; C=O, MEGA-8 = 1.231 (4), MEGA-11 = 1.162 (5) Å; C–N, MEGA-8 = 1.346 (4), MEGA-11 = 1.384 (7) Å. Since there is no great difference in the thermal-motion parameters, these differences are associated with the difference in the hydrogen bonding. In MEGA-8, the carbonyl O atom accepts a strong OH...O=C bond of 1.74 Å, whereas, in MEGA-11, the bond length is 1.94 Å. *Ab initio* molecular-orbital calculations on the formamide dimer by Jeffrey, Ruble, McMullan, DeFrees & Pople (1981) indicated that hydrogen-bond formation lengthened the C=O bond by 0.018 Å and shortened the adjacent C–N bond by 0.023 Å. The differences observed in these structures are in the same direction, but somewhat larger than calculated for the isolated formamide dimer.

The molecular packing is shown in Fig. 2. That of MEGA-8 is a head-to-head bilayer with interdigitizing

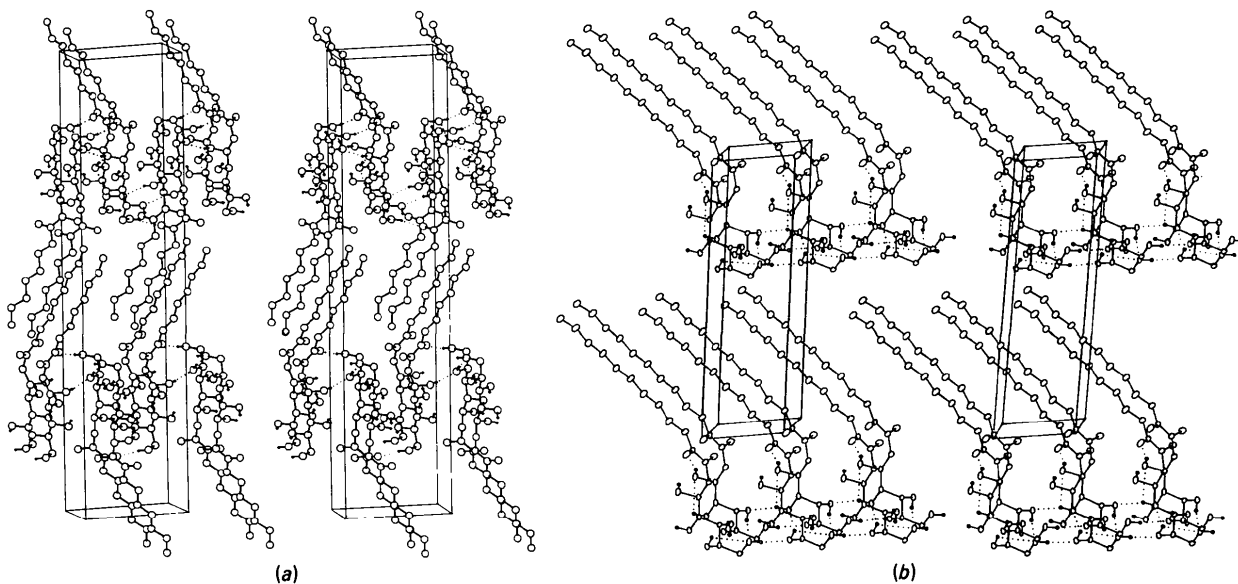


Fig. 2. Molecular packing in the crystal structures. (a) Head-to-head bilayer in MEGA-8; *c* vertical, *b* horizontal. (b) Head-to-tail monolayer in MEGA-11; *c* vertical, *b* horizontal.

alkyl chains similar to that observed in the crystal structures of the *n*-octyl α -glucopyranoside hemi- and monohydrates (Jeffrey *et al.*, 1987) and other *n*-alkyl pyranosides (Jeffrey, 1986). That of MEGA-11 is a head-to-tail monolayer with interdigitizing alkyl chains and, as expected, is isostructural with that of MEGA-9. This head-to-tail monolayer packing is also found in the crystal structure of *N*-(*n*-octyl)-*D*-gluconamide (Bhattacharjee *et al.*, 1985; * Zabel *et al.*, 1986), *N*-(*n*-heptyl)- and *N*-(*n*-decyl)-*D*-gluconamide (Müller-Fahrnow *et al.*, 1988) and *n*-decylribonamide (Baeyens-Volant, Fornasier, Szalai & David, 1986).

The hydrogen bonding

In both crystal structures, all six hydroxyl groups and the carbonyl O atoms are involved in the hydrogen bonding. The hydrogen-bonding repeat patterns are shown in Fig. 3.

In MEGA-8 there is a finite chain which originates at O4—H and terminates at the carbonyl O7. O2—H and

* In this paper, the structure was incorrectly identified as the gluconate.

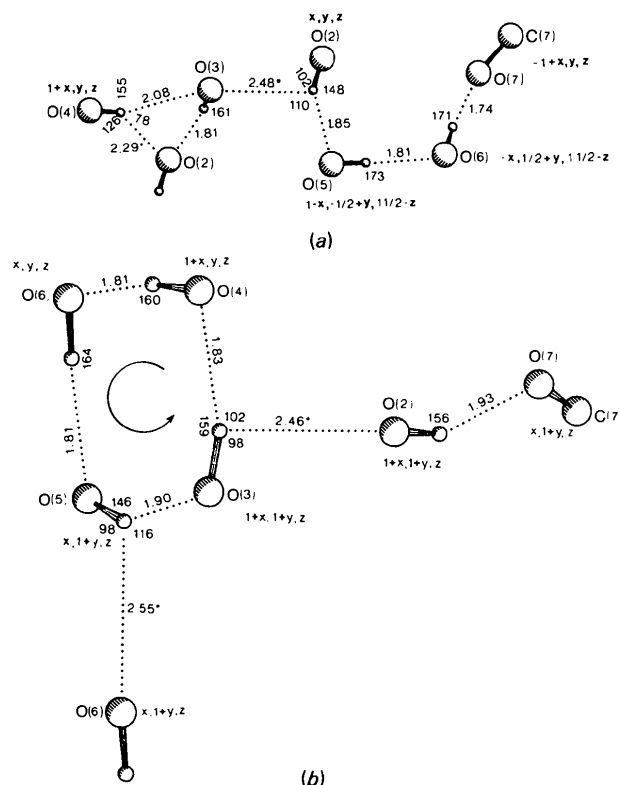


Fig. 3. Hydrogen bonding. (a) Finite chain pattern in MEGA-8. O(4)—H...O(2) intramolecular, O(2)—H...O(3) intramolecular. (b) Homodromic cycle; single link, weak intramolecular interaction in MEGA-11. The hydrogen-bond lengths are corrected for the effect of the O—H charge distribution by normalizing the covalent-bond lengths to the mean neutron diffraction value of 0.97 Å (Jeffrey & Lewis, 1978; Allen, 1984).

O4—H are three-center bonds, the minor components of which are intramolecular. That from O4—H...O2 is a stabilizing factor for the straight-chain glucitol conformation.

In MEGA-11, there is a homodromic cycle (Saenger, 1979) of strong bonds and a single O2—H...O7=C bond. These are linked by weak intramolecular interactions between the vicinal hydroxyl groups O5—H...O6, O3—H...O2. This is much the stronger hydrogen-bonding system of the two. There is a corresponding difference in the melting points, of 30 K, and in the solubility. As in mucic acid (*D*-galactaric acid; Jeffrey & Wood, 1982), the strong hydrogen bonding in the crystal results in insolubility in cold water and MEGA-11 is almost insoluble in water at room temperature.

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Some New Multi-Symmetric Packings of Equal Circles on a 2-Sphere

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Abstract

In an earlier paper [Tarnai & Gáspár (1987). *Acta Cryst.* **A43**, 612–616] four packing sequences were investigated to find locally extremal arrangements of equal circles on a sphere in tetrahedral, octahedral and icosahedral symmetry. In this paper one of these sequences is continued to find relatively dense arrangements in the case of large numbers of circles ($n = 150, 216, 300, 432, 750, 1080$).

Introduction

The Tammes problem is as follows: how must n equal non-overlapping circles be packed on a sphere so that the angular diameter of the circles will be as great as possible? With the analogy of the capsid structure of small 'spherical' viruses, Tarnai & Gáspár (1987) constructed some locally extremal arrangements in tetrahedral, octahedral and icosahedral symmetry. Regular tessellations were considered on these regular polyhedra. After Coxeter (1972) these tessellations were denoted by the symbol $\{3, q+\}_{b, c}$ where the number 3 means that the tessellation consists of equilateral triangles and the notation $q+$ refers to the fact that q or more than q triangles meet at the vertices of the tessellation. The suffixes b, c denote the coordination numbers of triangulation.

Four sequences of circle packings were investigated for $q = 3, 4, 5$: the tessellations $\{3, q+\}_{c+1, c}$ and $\{3, q+\}_{c+2, c}$ by removal and preservation of the vertices of the regular polyhedra $\{3, q\}$. For the calculations a procedure has been developed based on the 'heating technique' (Tarnai & Gáspár, 1983) considering the graph as a spherical bar and joint structure.

The first three ($c = 1, 2, 3$) elements of the four sequences were calculated by Tarnai & Gáspár (1987). The second sequence, the tessellation $\{3, q+\}_{c+2, c}$ with removal of the vertices of the base polyhedron, showed a strong regularity: for every c the topology of the subgraph on a face of the regular polyhedron was the same in the case of the tetrahedron, octahedron and icosahedron, and all of the subgraphs consist only of quadrangles with the exception of the middle of the face and the neighbourhood of the vertices where there are triangles. This regularity led us to continue this sequence to find relatively dense packings for large numbers of circles.

New results

In the case of $c = 4$ the regularity of the packing remained. The subgraph of these packings can be seen in Fig. 1 in a schematic form where (and similarly in Fig. 2) each great equilateral triangle composed of dashed lines is a face of the regular tetrahedron or