Colloid & Polym Sci 277:694–700 (1999)

SHORT COMMUNICATION

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Polymorphism of racemic 1,2-diols with aliphatic chains: isostructural series of phases in dry and hydrated samples

Received: 24 June 1998
Accepted in revised form: 15 February 1999

Dedicated to Prof. Dr. H. W. Meyer, Jena, on the occasion of his 65th birthday

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Abstract The different phases occurring in homologous dried *n*-alkane-1,2-diols (1,2-diols) and *n*-alkane-1,2,1′,2′-tetraols (1,2-boladiols), as well as in hydrated *n*-alkane-1,2-diols were characterised according to their X-ray diffraction patterns. The analysis of the X-ray data is mainly based on the small-angle region. The scattering in the wide-angle region, which may be regarded as fingerprint, is at present used for a qualitative discrimination of the phases. A homologous series of compounds is considered isostructural if the data show a linear relationship between the repeat distance d_L and the number of C atoms (C*m*) in a chain. Assuming that a given lateral packing remains constant, two characteristic values can be obtained from the plot of d_L versus C*m*. The slope *b* contains information about the tilt angle of the chains for the respective layer model and the intercept *a* represents the thickness of the hydrophilic part of the aggregate. A great variety of headgroup thicknesses and tiltings of the chains were found. Bilayers or double bilayers are the repeat unit in 1,2-diols, whereas in 1,2-boladiols the molecules are packed in monolayers. Water changes the packing of the chains and new packing modes with crossed chain axes were obtained. The ability to form hydrogen-bridged networks on both ends of the molecules forces the chains to occupy larger areas in dried 1,2-boladiols compared to the corresponding 1,2-diols.

Key words X-ray diffraction – Polymorphism – Diol derivatives – Liquid crystals – Hydrogen-bridged network

Introduction

The packing in two-dimensional (monolayers) and three-dimensional (bulk) systems of amphiphiles is determined by the competitive interactions between the hydrophilic headgroups and the hydrophobic tails. To understand these interactions at the atomic level it may be helpful to go back to such model compounds as 1,2-diols. They, for instance, are the most simple long-chain compounds which show a lamellar liquid-crystalline phase in the presence of water [1]. Aliphatic 1,2-diols exhibit both thermotropic and lyotropic liquid-crystalline behaviour [2] which distinguishes these substances from conventional amphiphiles.

The aim of these X-ray diffraction experiments is to characterise the different phases occurring in dried 1,2-diols and 1,2-boladiols, as well as in hydrated 1,2-diols. Boladiols are compounds which contain two hydrophilic headgroups located at opposite ends of the hydrophobic part of the molecule. They can be used as simple models for the chemically more complicated bolaamphiphilic lipids from archaebacteria membranes [3]. Additionally, boladiols are interesting on their own as they represent a new class of self-aggregating compounds which could have importance in material science [4, 5].

The initial results were confusing because of the unusual diffraction patterns which could not be explained by subcell diffraction or diffraction of known thermotropic or lyotropic phases [6–9]. Structural investigations of monolayers transferred on Formvar using transmission electron diffraction also show unusual packings even for aliphatic 1,2-diols [10]. The combination of the data from a homologous series which forms an isostructural series [1, 11] provides the possibility to classify the results and to derive parameters which are useful for further modelling.

Experimental

The substances investigated were kindly synthesised by the group of C. Tschierske at the Institute of Organic Chemistry, Martin Luther University Halle/Wittenberg. The chemical structures of the *n*-alkane-1,2-diols (1,2-diols) and the *n*-alkane-1,2,1′,2′-tetraols (1,2-boladiols) are shown in Fig. 1. C*m*-D ($m = n - 2$) and C*m*-BoD ($m = n - 4$) are used as abbreviations for the derivatives with *n* as the total number of C atoms of the molecules and *m* as the number of C atoms in the chains of the 1,2 diols and 1,2-boladiols, respectively, bearing in mind that each C atom with a hydroxyl group is involved in the headgroup of the amphiphilic compounds. Vacuum-dried samples and mixtures with 40 wt% water were investigated. The samples were sealed in thin-walled capillaries.

X-ray diffraction was performed with a HZG 4 powder diffractometer (Präzisionsmechanik Freiberg) using a transmission technique with Cu Kα radiation. The patterns are recorded as the difference between counts from balanced Ni and Co filters for the purpose of monochromatisation. For the calculations the limits of error are standard error for slope $< 5 \times 10^{-3}$ nm, standard error for intercept < 0.2 nm, correlation coefficient > 0.999.

The notation of the different phases is based on the superstructure, deduced from the long spacings (L-layer) and the state of the chains (r-rigid, f-fluid). Further distinctions are made with integers *X* (e.g., Lr2). Cr*X* denotes the crystalline state.

Results and discussion

Analysis

The analysis is mainly based on the small-angle region of the X-ray diffraction patterns of homologous 1,2-diols and 1,2-boladiols. From the long spacings a repeat distance d_L is calculated. The layers of one-dimensional packed aggregates consist of headgroups, hydrocarbon chains and, if present, water. A homologous series of compounds is considered isostructural if the data show a linear relationship between the repeat distance d_L and the number of C atoms (C*m*) in a chain [1]. If one accepts from the plot of d_L versus C*m* that a given lateral packing remains constant even as the chain length of the molecules changes within the range investigated, a formal extrapolation to C*m* = 0 yields two characteristic values. The slope *b* describes the difference in layer thickness as a result of changing the chain length by one C atom. For example, in a rigid hydrocarbon chain in the all-trans configuration a $\Delta_{C\text{-}C}$ value of 0.1257 nm can be estimated using the C-C bond length of 0.154 nm and the bond angle of 109.47°. Therefore a theoretical slope of $b = 0.2514$ nm is expected in a normal bilayer with perpendicular arranged chains. Usually, a value $b_{max} = 0.254$ nm which agrees very well with the averaged subcell constant c_s from single crystal studies [7] is used for further calculations [1]. Deviations from this value to lower or higher values point to the fact that the chains are tilted within the layers or that a different layer model has to be used, respectively. The tilt angle ϕ can be calculated according to $\cos \phi = b/(N\ \Delta_{C\text{-}C})$ which depends on the model used, for example, in monolayers $N = 1$, in bilayers $N = 2$ and in double bilayers $N = 4$ (Fig. 2A). In Table 1 models are presented concerning the layer packing of amphiphilic compounds. Most of them are the result of X-ray single-crystal studies and are also relevant in the powder-pattern studies.

The extrapolation to C*m* = 0 yields the intercept *a* which represents the thickness of the hydrophilic part of the aggregate and which is a characteristic value only within the range investigated. The division of *a* by an integer *I* gives the thickness of the hydrophilic part of a single molecule, where the integer *I* also depends on the model used (Table 1). For example, $I = 2$ is used for a bilayer with oppositely arranged molecules as well as for a double bilayer with interdigitated headgroups or a monolayer consisting of bolaamphiphiles (Fig. 2B).

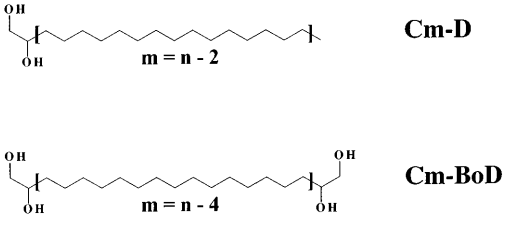

Fig. 1 Chemical structures of 1,2-diols and 1,2-boladiols and the abbreviations used. *n* is the total number of C atoms of the molecule and *m* is that of the chain

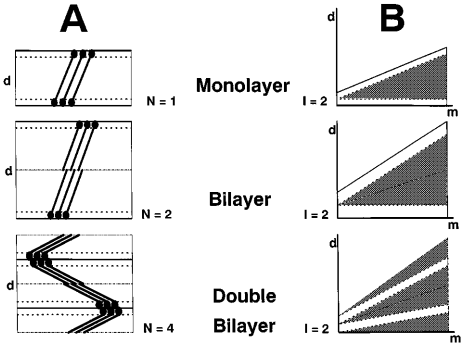

Fig. 2A, B Structural models of the phases investigated. **A** Molecular packing within different aggregates illustrating the sense of the integer *N*. **B** Interpretation of the isostructural series with models with $I = 2$ but with different values of *N* (See Table 1)

Table 1 Models of layered structures with different arrangements of headgroups and chains for single-chain amphiphiles. (*N*: number of layers within the repeat distance; *I*: integer to divide the hydrophilic part within the repeat distance into a molecular headgroup thickness; exceptions from the head-to-head packing are noted)

N	Layer type	*I*	Headgroups	Chains	Examples
1	Monolayer	1	–	–	So far not observed: head-to-tail
1	Monolayer	1	Interdigitated	–	So far not observed: bolaamphiphile
1	Monolayer	2	Adjacent	–	Fig. 2A, B, Ref. [23]
1	Monolayer	1	Interdigitated	Interdigitated	[24] (PPC, HPC, $PPEM_2$)
1	Monolayer	2	Adjacent	Interdigitated	[24] (deoxy PC)
2	Bilayer	2	–	–	head-to-tail: Ref. [25]
2	Bilayer	2	Interdigitated	–	[26]
2	Bilayer	4	Adjacent	–	[27]
2	Bilayer	1	Interdigitated	Adjacent	So far not observed
2	Bilayer	2	Adjacent	Adjacent	Fig. 2A, B, Ref. [28] (deoxy HPA1, LPA^{2-}, PPE)
2	Bilayer	2	Interdigitated	Interdigitated	[28] (OMPC)
2	Bilayer	4	Adjacent	Interdigitated	[28] (Sp, TASp, Glc-PSp)
4	Double bilayer	2	Interdigitated	Adjacent	Fig. 2B, Ref. [29]
4	Double bilayer	4	Adjacent	Adjacent	Fig. 2A, Ref. [30] (deoxy HPA2)

From this data additional parameters can be calculated. The area per molecule, S, within the surface is derived according to $S = \Sigma/\cos\phi$. The cross section Σ of an aliphatic chain in the lowest energy configuration amounts to 0.185 nm^2 for molecules packed with parallel long axes. The single crystal data of *n*-heptane-1,2,3-triol allow the evaluation of Σ in the case of crossed chain axes. With $S = 0.2185$ nm^2 [12] and a recalculated tilt angle $\phi = 14°$ a value of $\Sigma = 0.212$ nm^2 was estimated. Note that the high value of this apparent cross section corresponds to a molecule conformation as a straight line without a trans–gauche–trans kink between head and tail and is realised in a structure which is stabilised by a hydrogen-bridged network between the headgroups.

One has to keep in mind that the use of these values from crystalline phases only allows a rough estimation of the area S in mesomorphic phases, for example, the area per chain is typically 0.196 nm^2 in orthogonal rotator phases of *n*-paraffins [13]. Therefore, the value of S can be underestimated in mesomorphic phases. The solution of this problem is the individual indexing of the spacings in the wide-angle region of the phase investigated which allows the determination of the lattice constants as well as the cross section of the chain packing.

Finally, verification of the structural model is possible by calculating the packing coefficient k introduced by Kitaigorodski [14]. The idea in the case of mesomorphic phases of amphiphiles is to define a unit cell V_U in the layer structure according to an assumed model using the experimental data d_L and S. In the case of single-chain amphiphiles arranged in a bilayer ($N = 2$) with adjacent headgroups ($I = 2$) V_U, for instance, is given according to $V_U = d_L S$ with two molecules within this cell. Taking into consideration the van der Waals volume V_{vdW} of the molecules included, the packing coefficient k is defined according to $k = 100\%\ V_{vdW}/V_U$. The V_{vdW} values of homologous 1,2-diols and 1,2-boladiols are calculated according to the rules and with the values of Kitaigorodski [14] using $V_{vdW} = [78.696 + (Cm-1) \times 16.8169] \times 10^{-3}$ nm^3 and $V_{vdW} = (112.3554 + Cm \times 16.8169) \times 10^{-3}$ nm^3, respectively. Physically reasonable values for k are reported as $k = 65$–77% for crystals, $k < 60\%$ for glasses, $k < 58\%$ for liquids and $k < 50\%$ for gases [15].

No extended analysis of the wide-angle X-ray scattering was performed. The experimental data are presented for comparison and discussed as the fingerprint of the respective phases only qualitatively.

Phase Cr1 of dry 1,2-diols

The structure Cr1 occurs in dry, melt-crystallised samples of 1,2-diols. With this thermal history they are comparable with second heating runs in differential scanning calorimetry experiments.

Examples of the X-ray diffraction patterns are given in Fig. 3 (left) for C10-D and C14-D, respectively. The scattering in the wide-angle region, which may be regarded as its fingerprint, shows many sharp reflections which are typical for a crystalline phase. The three predominant reflections as well as the weaker peaks surrounding them are similar for both homologous compounds despite their different long spacings.

The data of this isostructural series were analysed on the basis of the bilayer model schematically sketched in Fig. 3 (right). A headgroup thickness of 0.45 nm and a tilt angle of 19° were calculated for the chains. Using an area per molecule of $S = 0.196$ nm^2 a packing coefficient of $k = 71\%$ is derived (Table 2) which represents the physical state of Cr1 quite well and verifies the model used.

Phase Lr3 of dry 1,2-diols

The phase Lr3 appears as a monotropic phase on cooling the short-chain homologues ($9 < Cm < 14$) of 1,2-diols. Its typical X-ray diffraction pattern is shown in Fig. 4 (left). The short spacings are similar to those of the phase Lβ' in 1,2-boladiols [16] (see Fig. 7, left), but until now a satisfactory indexing has not been available.

In Fig. 4 (right) the isostructural series of the phase Lr3 and a schematic model of the C15-D homologue are shown. From the linear regression (Table 2) a thickness of the headgroups of $L_H = 0.50$ nm is derived. For the tilt angle of the chains a value of $\phi = 27°$ is calculated based on a bilayer model with oppositely arranged molecules. Each molecule occupies an area of $S = 0.217$ nm^2. The packing coefficient of $k = 68\%$ is at the lower limit for crystals.

A structural model proposed by van Doren et al. [17] explained the shortening of the repeat distance with respect to the molecule length as an interdigitation of the diol headgroups; however, their model did not take into consideration a possible tilting of the chains. The given value of the tilt angle is the result of an analysis within the isostructural series which does not require any assumptions.

Phase Cr3 of hydrated 1,2-diols

Mixtures of 1,2-diols with excess water show the low-temperature phase Cr3. It is known from systematic hydration experiments that the mesomorphic phase occurring at higher temperatures squeezes out water during the phase transition to the low-temperature phase [18]. From the first appearance of the eutectic temperature in the binary phase diagram it has been concluded that the headgroups contains only a minimal amount of water in the phase Cr3 [6, 19].

Selected X-ray diffraction patterns are shown in Fig. 5 (left). The appearance of strong and sharp small-angle spacings especially for the C13-D sample is remarkable. In addition, only two strong reflections

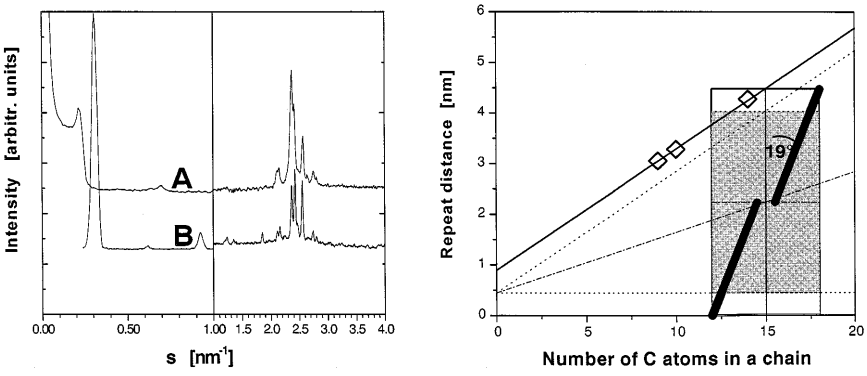

Fig. 3 *Left*: X-ray diffraction patterns of the phase Cr1. *A*: C14-D (25 °C, dry); *B*: C10-D (25 °C, dry). *Right*: isostructural series of phase Cr1 in homologous dry 1,2-diols with a structural model for the C15-D homologues

Table 2 Analysis of the isostructural series of *n*-alkane-1,2-diols (C*m*-D) and *n*-alkane-1,2,1′2′-tetraols (C*m*-BoD). (L_H: headgroup thickness; ϕ: tilt angle of the chains; S: area per molecule in the surface; k: packing coefficient)

Phase Sample history	Linear regression $d_L = a + b \times Cm$	L_H (nm)	ϕ	S (nm^2)	k (%)
C*m*-D, dried					
Cr1 melt-crystallised	$d_L = 0.8936 + 0.2395 \times Cm$ Bilayer	0.45	19°	0.196	71
Lr3 monotropic mesomorphic	$d_L = 0.9985 + 0.2251 \times Cm$ Bilayer	0.50	27°	0.208	68
C*m*-D, fully hydrated					
Cr3 crystallised from aqueous dispersion	$d_L = 0.3233 + 0.3037 \times Cm$ Double bilayer	0.16	53°	0.309	79
Lr4 mesomorphic	$d_L = 1.6167 + 0.2433 \times Cm$ Bilayer, crossed chain axes	0.62 (diol) 0.18 (water)	16°	0.221	61
Lf1 liquid crystalline	$d_L = 2.1914 + 0.1586 \times Cm$ Bilayer	1.10 (diol + water)	–	–	–
C*m*-BoD, dried					
Cr2 melt-crystallised	$d_L = 0.5565 + 0.1049 \times Cm$ Monolayer	0.28	34°	0.223	77%
Lr1 mesomorphic	$d_L = 0.7729 + 0.1036 \times Cm$ Monolayer	0.38	35°	0.226	69%
Lβ' [16] mesomorphic	$d_L = 0.6704 + 0.1107 \times Cm$ Monolayer	0.34[a]	29°[a]	0.212[a]	74%[a]
Lα [16] liquid crystalline	$d_L = 0.6729 + 0.0981 \times Cm$ Monolayer	0.32[a]	–	–	–

[a] Recalculated from Ref. [16]

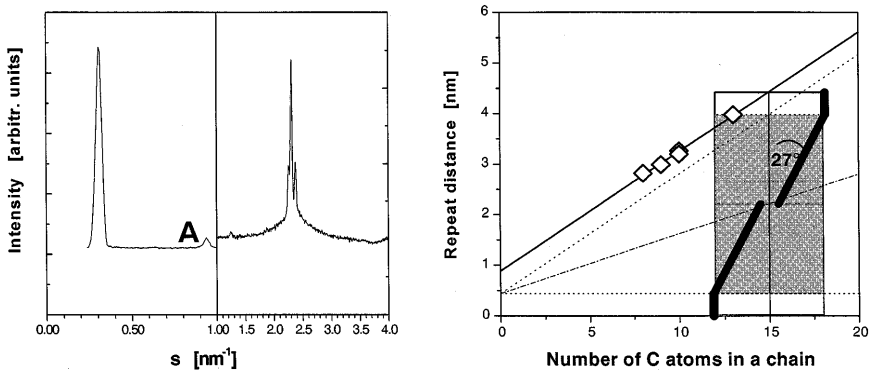

Fig. 4 *Left*: X-ray diffraction pattern of the phase Lr3. *A*: C10-D (45 °C, dry). *Right*: isostructural series of phase Lr3 in homologous dry 1,2-diols with a structural model for the C15-D homologue. Values for C8-D, C9-D and C10-D adopted from Ref. [17]

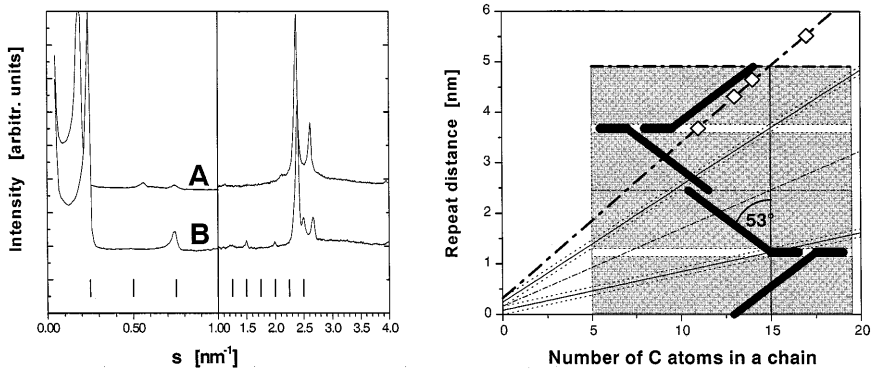

Fig. 5 *Left*: X-ray diffraction patterns of the phase Cr3. *A*: C17-D (−6.9 °C, hydrated); *B*: C13-D (−5.3 °C, hydrated, 10 orders of the long spacing are marked). *Right*: isostructural series of phase Cr3 in homologous hydrated 1,2-diols with a structural model for the C15-D homologue

are found in the wide-angle region. Usually this pattern is interpreted as the scattering from an orthorhombic subcell $O_{\perp}$. The finding indicates that the low-temperature phase in hydrated 1,2-diols has a structure which can be considered as a subgel phase, usually found in lipid/water systems, rather than a crystalline phase.

The analysis of the isostructural series of Cr3 gives very surprising results (Table 2). The slope b is so large that it exceeds the maximum value for a bilayer model (b_{max} = 0.254 nm); therefore a double bilayer model is proposed (Fig. 5, right) which yields an extreme tilt angle of 53°. Additionally, an extremely small thickness for the headgroups (L_H = 0.162 nm) is extrapolated based on this model. It is assumed that the headgroups of adjacent surfaces are interdigitated and aligned parallel to the interface in order to explain such a small value. The area per molecule of S = 0.309 nm^2, calculated from the large tilt angle under the assumption of parallel packing of the chain axes within the bilayer, correlates well with the proposed headgroup conformation; however, the verification of the structural model using the packing coefficient is complicated due to the difference between a normal double bilayer model (N = 4, I = 4, Fig. 2A, bottom) and a double bilayer model with interdigitated headgroups (N = 4, I = 2, Fig. 2B, bottom). Four complete molecules are included in the calculation of k for the double bilayer model. As we propose interdigitation of the headgroups for Cr3 (Fig. 5, right), the measured d_L value reflects the contribution of four chain sublayers but only two sublayers of headgroups, and therefore the packing coefficient is too high. A correction is introduced as an additional headgroup contribution. The speculative term $d_L = 2a + b\,Cm$ for the Cr3 homologous series using experimentally estimated parameters a and b, agrees with the molecule number used for the calculation and yields a packing coefficient of k = 79%. This value is at the limit for dense-sphere packing. On the other hand, each contribution of rotational freedom in mesomorphic packings increases the cross section Σ of the chains (0.196 nm^2) and decreases k for this model (k = 75%).

Phase Lr4 of hydrated 1,2-diols

Lr4 is the high-temperature mesomorphic phase in hydrated 1,2-diols. It appears as a monotropic phase in short-chain homologues but becomes enantiotropic at chain lengths above C13. Most of the diffraction experiments were performed in the monotropic supercooled phase. The X-ray diffraction patterns show the peculiarity that the profile of the spacings in the wide-angle region changes continuously with increasing chain length (Fig. 6, left).

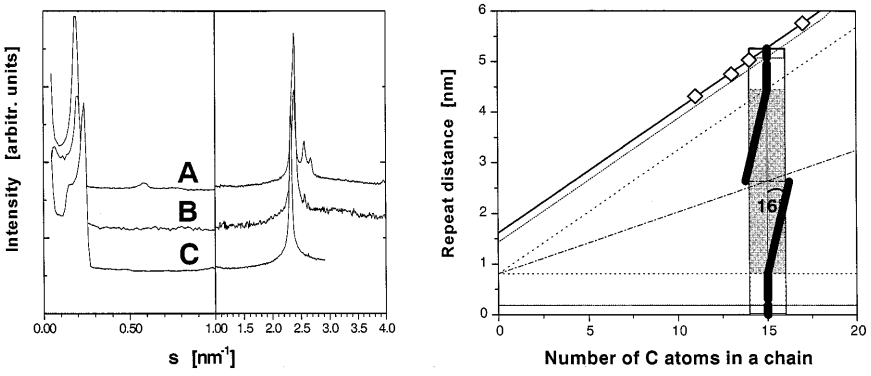

Fig. 6 *Left*: X-ray diffraction patterns of the phase Lr4. *A*: C17-D (0 °C, hydrated); *B*: C14-D (25 °C, hydrated); *C*: C11-D (25 °C, hydrated). *Right*: isostructural series of phase Lr4 in homologous hydrated 1,2-diols with a structural model for the C15-D homologue

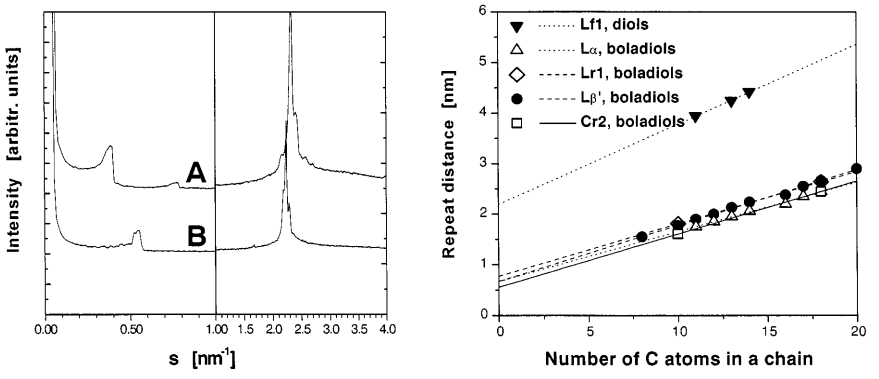

Fig. 7 *Left*: X-ray diffraction patterns of the phase Lr1. *A*: C18-BoD (97 °C, dry); *B*: C10-BoD (110 °C, dry). *Right*: isostructural series of phase Lf1 in homologous hydrated 1,2-diols in comparison to the approximate straight lines for the phases Cr2 and Lr1 in dried 1,2-boladiols (Lα and Lβ' adopted from Ref. [16] are included)

The extrapolation within the isostructural series (Fig. 6, right) gives a very high value for the thickness of the hydrophilic part and a small value for the tilt angle (ϕ = 16°), which yields an area per molecule of S = 0.221 nm^2. As the amount of bound water in this phase was found to be n_w = 2.5 water molecules per headgroup [1, 18, 19] the hydrophilic region has to be divided into two parts: that of bound water and that of the diol headgroup. According to the results of a single-crystal study of a 1,2,3-triol [12] a model for Lr4 is proposed which assumes slightly crossed chain axes filling the space below the diol headgroups which build a lateral two-dimensional hydrogen-bridged network. The bound water is arranged in a separate layer. Its thickness L_w = 0.184 nm can be derived according to

$$L_w = n_w M_w / N S N_A \times 10^{-21}\,\text{nm}\ , \quad (1)$$

where n_w is the number of water molecules per headgroup, M_w is the molar mass of water, N is an integer according to the model used (bilayer: N = 2) and N_A is Avogadro's number) [20]. Then, for the diol headgroup a thickness L_D remains which represents the most extended headgroup conformation in comparison with other mesomorphic phases (Table 2). The packing coefficient k = 61% has a physically reasonable value.

Phase Lf1 of hydrated 1,2-diols

In hydrated 1,2-diols a liquid-crystalline lamellar phase Lf1 is found in which the chains are fluid. Surprisingly a straight line in the isostructural series was also observed in the liquid-crystalline phase (Fig. 7, right). This result seems to be in contrast to the literature which states that the increase in the repeat distance decreases with increasing chain length in a homologous series [21]; however, our results are confirmed by the observation of a linear relationship for d_L versus Cm found for similar substances [22]. This allows the extrapolation to zero chain length even in the case of a liquid-crystalline phase of hydrated 1,2-diols and the determination of the headgroup thickness equilibrated with water. Keeping in mind that the hydration limit in the phase Lf1 is nearly the same as in the mesophase Lr4, this thickness is very large; therefore, one has to assume that in addition to the diol headgroup and bound water a certain amount of trapped water must also be included. This type of water changes the bilayer distance but does not influence the calorimetric data. Such behaviour has already been observed in lyotropic phases of lipid systems [20].

Isostructural series of dried 1,2-boladiols

For 1,2-boladiol systems only preliminary data are available based on investigations of the C10-BoD and C18-BoD derivatives, respectively. The analysis of the isostructural series of the phases Cr2 and Lr1 results in a monolayer model. In comparison with the 1,2-diols, the headgroups exhibit only a small thickness and the chains are more tilted (Table 2).

The fingerprint pattern of the mesomorphic phase Lr1 (Fig. 7, left) is different to that of Lβ' given in the literature [16]. This experimental result indicates that different polymorphism may exist in 1,2-boladiols depending on the history of the sample.

Conclusions

The characterisation of phases according to their diffraction patterns and the analysis of the isostructural series are useful for describing homologous 1,2-diols and 1,2-boladiols.

Dried 1,2-diols form phase structures with extended headgroups and relatively small tilt angles of the chains. In the presence of water different structures are built in the crystalline state as well as in the mesomorphic state. The analysis of the hydrated phase gives an indication that chain packings with crossed chain axes are also stable in a mesomorphic phase. Despite the water bound to the headgroups, trapped water arranged between the headgroups was also observed in the liquid-crystalline phase. This type of water does not influence the thermal parameters. Dried 1,2-boladiols form monolayers. Nevertheless, a smaller headgroup thickness and larger chain tilt angle were found compared to the 1,2-diols. This shows that the presence of headgroups on both ends of the chains forces the chains to occupy a larger area per molecule within the surface.

The estimated quantities such as tilt angle and headgroup thickness demonstrate a remarkable variability of the packing of diols. Two prerequisites are given in this homologous series; namely a conformational flexibility between the headgroup and the chain and strong attractive interactions mainly in the lateral direction. The experimental results imply that different possibilities exist to form two-dimensional hydrogen-bridged networks. These networks are also the basic features for the stabilisation of the liquid-crystalline lamellar structure in 1,2-diol/water mixtures which was not observed in other long-chain compounds of comparable simple chemical structure (paraffins, fatty acids).

Acknowledgements Financial assistance from the Deutsche Forschungsgemeinschaft (SFB 197) is gratefully acknowledged. We also thank Petra Hartmann for technical assistance and Christine DeWolf for critical reading of the manuscript.

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