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Heat capacities near room temperature of ten solid alkane- α , ω -diols HO–(CH₂)_n–OH where n = 6 and $8 \le n \le 16$

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The molar heat capacities at constant pressure $C_{p,m}$ of a homologous series of alkane- α , ω -diols HO–(CH₂)_n–OH, where *n* has values of 6 and (8 to 16), were determined by using differential scanning calorimetry over the temperature range (280 to 310) K, in flowing gas. The measured values of $C_{p,m}$ showed a very good linear dependence on temperature, except 1,9-nonanediol and 1,13-tridecanediol. The heat capacities at T = 298.15 K were obtained by interpolating the smoothed fitting equations as a function of temperature. The contribution to the heat capacity at T = 298.15 K of the CH₂ group was derived from the slope of the $C_{p,m}$ values of diols as a function of the number of carbon atoms in their molecules, and compared with the values obtained from a comprehensive collection of literature data for homologous alkyl organic compounds in the solid state. ©1999 Academic Press

KEYWORDS: alkane- α , ω -diols; methylene group; heat capacity; differential scanning calorimetry

1. Introduction

Simple organic compounds, including alkanediols,⁽¹⁾ are useful model systems for solute–solvent interactions in dilute solution. However, of the data needed to obtain a comprehensive thermodynamic description of the process, molar heat capacities $C_{p,m}$ of pure solutes are often missing from the literature. These quantities are employed to convert to T = 298.15 K,

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both enthalpies of vaporization or sublimation measured at other temperatures for liquid or solid solutes, and hence derive their standard enthalpies of solvation⁽²⁾ by using enthalpies of solution at infinite dilution. They can also be employed to obtain partial molar heat capacities of solutes through addition to the changes of heat capacity due to solution processes at infinite dilution.⁽³⁾

As a part of our study on the thermodynamic properties of a number of series of organic compounds,^(4–15) this work reports the molar heat capacities measured by using differential scanning calorimetry (d.s.c.) near room temperature of ten solid alkane- α , ω -diols, with the general formula HO–(CH₂)_n–OH, namely 1,6-hexanediol, and 1,8-octanediol to 1,16-hexadecanediol. To our knowledge, no previous heat capacities of solid terminal diols at T = 298.15 K are available in the literature, except for 1,6-hexanediol⁽¹⁶⁾ and 1,10-decanediol.⁽¹⁷⁾ Furthermore, these results contribute to an evaluation of the contribution to the heat capacity of the CH₂ group in solid alkyl compounds.

2. Experimental

The alkane- α , ω -diols investigated were analytical grade reagents from various suppliers. Original mass fraction purities (when indicated) ranged from 0.98 to 0.99, and are listed in table 1. Only those terminal diols that are solid at room temperature were considered in this work, from N(C) = 6 to N(C) = 16, where N(C) denotes the number of C atoms in the molecule, but 1,7-heptanediol N(C) = 7 was omitted because it melts at $T \approx 290$ K. All commercial products were purified by successive crystallizations from (water + ethanol) or absolute ethanol solutions, except 1,14-tetradecanediol, which was used as received. Since 1,13-tridecanediol is not commercially available, we prepared this compound in our laboratory by reduction of 1,11-undecandicarboxylic acid (Pfaltz & Bauer) by LiAlH₄ (Aldrich, powder, mass fraction 0.95) in a tetrahydrofuran (Aldrich, anhydrous, mass fraction 0.999) solution. The precipitated compound was then purified by up to seven crystallizations from absolute ethanol and repeated sublimations.

All the samples were subjected to prolonged drying in a vacuum oven and in most cases the final purification of the samples also included sublimation under vacuum. Purities were determined by the d.s.c. peak profile method^(18, 19) and are reported in the last column of table 1. The procedures adopted significantly increased the purity despite the well-known difficulty of purifying this class of compound. The final mass fraction of our samples was sometimes better than 0.995, but did not reach the excellent values we have obtained in other cases, *e.g.* for *N*-alkylureas.⁽¹³⁾

In the heat flow calibration procedure, high purity benzoic acid from NPL (SRM M16-06, mass fraction 0.9999) was used as reference material.

Heat capacities were measured by using d.s.c. in the temperature range (280 to 310) K with a Perkin–Elmer DSC-2 apparatus calibrated for temperature and enthalpy at both superambient and subambient temperatures.^(20, 21) However, this calibration procedure has been recently improved as described in reference 13. In particular: benzoic acid in the form of pressed pellets was used as a calorimetric standard over the whole experimental temperature range; a (0.10He + 0.90Ne) mixture (He mass fraction 0.999995 and Ne mass fraction > 0.9998) supplied by SIAD (Dalmine, Bergamo, Italy) was employed as a more

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Compound	$M/(g \cdot mol^{-1})^a$	Origin	w^b	w_{f}			
1,6-Hexanediol	118.176	Aldrich	>0.99	0.994			
1,8-Octanediol	146.230	Aldrich	0.98	0.985			
1,9-Nonanediol	160.257	Aldrich	0.98	0.996			
1,10-Decanediol	174.283	Aldrich	0.98	0.998			
1,11-Undecanediol	188.310	Pfaltz & Bauer		0.996			
1,12-Dodecanediol	202.337	Aldrich	0.99	0.995			
1,13-Tridecanediol	216.364	this work		0.993			
1,14-Tetradecanediol	230.391	Aldrich		0.988			
1,15-Pentadecanediol	244.418	Pfaltz & Bauer		0.996			
1,16-Hexadecanediol	258.445	Aldrich	0.98	0.991			

TABLE 1. List of the alkane- α , ω -diols, molar mass M, commercial origin, mass fraction w^b , and final mass fraction w_f determined in this work by the d.s.c. peak profile method^(18, 19)

^{*a*}Based on the 1991 IUPAC Table of Standard Atomic Weights of the Elements. ^{*b*} *w* was not indicated by the supplier for 1,11-undecanediol, 1,14-tetradecanediol, and 1,15-pentadecanediol, whereas 1,13-tridecanediol was prepared in this work. ^{*c*} Used as received.

appropriate gaseous thermal exchanger near room temperature; the samples were prepared in the form of pellets about 1 mm thick and 4 mm in diameter, and mass in the range (10 to 25)·10⁻³ g, stored over P₂O₅ in a vacuum desiccator at room temperature and then sealed in Al volatile-sample pans, in order to prevent any contact with moisture. A compromise was achieved between scanning rate, sample mass, and instrumental sensitivity range to obtain a sufficiently large signal-to-noise ratio.

The ordinate-displacement method was adopted and covered the temperature range (30 to 40) K. Two different reference samples and the empty sample holder were usually scanned before and after recording the series of runs necessary to explore the full temperature range for each sample of the studied compounds. The average value of the ordinate displacements in two or three subsequent runs was determined for each sample at the selected temperatures. For the reference samples and empty holder, the average of all the recorded values was taken into account.

The accuracy of the measurements was estimated to be within 2 per cent over the whole temperature range considered. The uncertainty of the temperature at the scanning rate of 0.167 K \cdot s⁻¹ selected was within \pm 0.2 K. More details concerning the calibration and experimental procedures adopted for measurements with our Perkin-Elmer DSC-2 equipment are reported in an earlier paper.⁽¹³⁾

No solid-to-solid transitions were found in the temperature range of the measurements. A complete investigation concerning solid-to-liquid and solid-to-solid transitions from T = 298 K for this series of terminal diols is being prepared for publication.



FIGURE 1. Molar heat capacities $C_{p,m}$ of alkane- α , ω -diols as a function of temperature T. a, 1,6-hexanediol; b, 1,8-octanediol; c, 1,10-decanediol; d, 1,12-dodecanediol; e, 1,14-tetradecanediol; f, 1,16-hexadecanediol.

3. Results and discussion

The results of the measurements are reported in table 2, where $C_{p,m}$ values are averaged over two or more experimental runs. They are also presented in figures 1 and 2 to illustrate their substantial linearity as a function of temperature, though for 1,13-tridecanediol, and partially for 1,9-nonanediol, a slight progressive increase of the slope was observed. No other determinations of heat capacities of terminal diols that are solid at room temperature are available in the literature, except for 1,6-hexanediol⁽¹⁶⁾ and 1,10-decanediol.⁽¹⁷⁾

are available in the literature, except for 1,6-hexanediol⁽¹⁶⁾ and 1,10-decanediol.⁽¹⁷⁾ The linear equations $C_{p,m}/(J \cdot K^{-1} \cdot mol^{-1}) = A + B\{(T/K) - 298.15)\}$ were obtained by a least-squares analysis of all the data, except that for 1,13-tridecanediol the values at



FIGURE 2. Molar heat capacities $C_{p,m}$ of alkane- α , ω -diols as a function of temperature T. a, 1,9-nonanediol; b, 1,11-undecanediol; c, 1,13-tridecanediol; d, 1,15-pentadecanediol.

just the first four temperatures were extrapolated to T = 298.15 K. The results are shown in table 3, where columns 2, 3, and 4 give the intercepts at T = 298.15 K (*A*), slopes (*B*), and product-moment correlation coefficients (*r*), respectively. The only two values in the literature^(16, 17) are in agreement with those obtained in the present work.

Figure 3 shows the heat capacities at T = 298.15 K (table 3, column 2) as a function of the number of carbon atoms of the ten alkane- α , ω -diols. Its slope represents the average contribution of the CH₂ group to the total heat capacity of various diols and gave a value of (23.05 ± 0.98) J·K⁻¹·mol⁻¹. Furthermore, if the heat capacity at T = 298.15 K for 1,6-hexanediol⁽¹⁶⁾ and that extrapolated to the same temperature from the solid 1,2-ethanediol⁽²²⁾ are also considered, the intercept at n = 0 gives (58.4 ± 6.6) J·K⁻¹·mol⁻¹

Т	п									
K	6	8	9	10	11	12	13	14	15	16
280	176.79	220.57	236.75	261.93	276.74	307.46	334.27	354.16	352.36	396.62
285	181.38	224.94	242.64	267.17	282.35	312.99	341.02	361.26	359.13	405.15
290	184.44	229.22	247.95	272.19	287.82	319.54	351.23	368.23	366.86	413.14
295	187.86	233.58	253.00	276.41	294.06	326.45	360.98	375.97	372.72	421.16
300	191.48	238.21	259.24	281.16	300.28	332.75	370.82	382.04	380.61	429.53
305	195.58	242.48	266.64	286.10	307.13	339.77	383.38	391.17	388.17	437.70
310	201.31	247.67	276.36	291.66	314.09	347.62	399.87	399.00	396.86	446.96

TABLE 2. Experimental molar heat capacities $C_{p,m}/(J \cdot K^{-1} \cdot mol^{-1})$ of the alkane- α , ω -diols HO–(CH₂)_n–OH at selected temperatures

TABLE 3. Molar heat capacities of the alkane- α , ω diols as a function of temperature from the equation $C_{p,\mathrm{m}}/(J \cdot \mathrm{K}^{-1} \cdot \mathrm{mol}^{-1}) = A + B\{(T/\mathrm{K}) - 298.15\}$ in the interval T = (280 to 310) K

	/	
A^{a}	B^a	r^b
190.7 ± 0.3^c	0.779 ± 0.031	0.99598
236.5 ± 0.1	0.896 ± 0.010	0.99966
258.7 ± 0.7	1.272 ± 0.066	0.99331
279.6 ± 0.1^d	0.972 ± 0.014	0.99949
298.4 ± 0.3	1.243 ± 0.025	0.99896
330.7 ± 0.3	1.338 ± 0.026	0.99909
365.8 ± 1.3	1.807 ± 0.110	0.99632
380.4 ± 0.3	1.487 ± 0.029	0.99908
378.2 ± 0.3	1.467 ± 0.030	0.99897
426.4 ± 0.2	1.661 ± 0.014	0.99981
	$\begin{array}{c} A^{a} \\ 190.7 \pm 0.3^{c} \\ 236.5 \pm 0.1 \\ 258.7 \pm 0.7 \\ 279.6 \pm 0.1^{d} \\ 298.4 \pm 0.3 \\ 330.7 \pm 0.3 \\ 365.8 \pm 1.3 \\ 380.4 \pm 0.3 \\ 378.2 \pm 0.3 \\ 426.4 \pm 0.2 \end{array}$	A^a B^a 190.7 \pm 0.3 ^c 0.779 \pm 0.031 236.5 \pm 0.1 0.896 \pm 0.010 258.7 \pm 0.7 1.272 \pm 0.066 279.6 \pm 0.1 ^d 0.972 \pm 0.014 298.4 \pm 0.3 1.243 \pm 0.025 330.7 \pm 0.3 1.338 \pm 0.026 365.8 \pm 1.3 1.807 \pm 0.110 380.4 \pm 0.3 1.487 \pm 0.029 378.2 \pm 0.3 1.467 \pm 0.030 426.4 \pm 0.2 1.661 \pm 0.014

^{*a*}Uncertainties are twice the standard deviation of the mean. ^{*b*}Product-moment correlation coefficient. ^{*c*} A = 203.60.⁽¹⁶⁾ ^{*d*} A = 270.10.⁽¹⁷⁾

corresponding to the molar heat capacity at T = 298.15 K of solid HO–OH, in excellent agreement with the value of (58.7 ± 0.1) J·K⁻¹·mol⁻¹ for H₂O₂(cr) at the same temperature, as extrapolated by a least-squares analysis from the data of Giguère *et al.*⁽²³⁾ in the subambient temperature interval (130 to 240) K.

In table 4, the two molar heat capacity contributions of the CH₂ group for our ten solid alkane- α , ω -diols derived (first line), or also including references 16 and 22 (second line) are compared with those obtained from a comprehensive collection of literature data embracing many homologous series of solid organic alkyl compounds. The values in column 4 (third line and following) were derived from the slopes of the plots of the smoothed $C_{p,m}$ values at T = 298.15 K as a function of the total number of carbon atoms in the molecules of each homologous series. All these values were recalculated from original data, and in many cases



FIGURE 3. Molar heat capacities $C_{p,m}$ of alkane- α , ω -diols at T = 298.15 K as a function of the number of C atoms N(C).

the temperature interval considered was restricted to obtain a sufficiently representative set of values near room temperature, which can be satisfactorily interpolated by a linear plot. Furthermore, the $C_{p,m}$ values of those compounds that are liquid at room temperature were obtained for solids at T = 298.15 K by extrapolating the original subambient temperature data by a least-squares analysis. The number N of compounds considered for each series, and the lowest and highest total carbon atoms in their molecular backbone N(C) are set out in columns 2 and 3. Differences between these two columns can originate either from the presence of isomers in a complete, or almost complete, series of homologous compounds (positive differences), or from the absence of reliable literature values at T = 298.15 K for some terms of the series (negative differences).

The contribution to the molar heat capacities calculated in this work for the CH₂ group in solids ranges from 20.6 $J \cdot K^{-1} \cdot mol^{-1}$ for alkanoic acids to 26.8 $J \cdot K^{-1} \cdot mol^{-1}$ for both *N*-alkylureas and *N*-acetylamino acid amides. The data reported in table 4 can be related to the type of crystal structures in which alkyl chains are packed in the series of solid organic homologous compounds considered.

In *n*-alkanes, dispersion forces alone are present, whereas in all the other series H-bond networks play a more or less significant role in building up crystal structures. The influence of the shared H-bonds depends on the nature and size of the functional groups, so that alkyl

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Compounds	Ν	$N(\mathbf{C})$	$C_{p,\mathrm{m}}/(\mathrm{J}\cdot\mathrm{K}^{-1}\cdot\mathrm{mol}^{-1})$	References
Alkane- α , ω -diols	10	6 to 16	23.05 ± 0.98	this work
Alkane- α , ω -diols	12	2 to 16	22.5 ± 0.6	this work, 16, 22
<i>n</i> -Alkanes	22	4 to 33	22.5 ± 0.4	24–26
n-Alkan-1-ols	13	1 to 16	22.9 ± 0.6	27-32
<i>n</i> -Alkanoic acids	12	7 to 20	20.6 ± 0.7	33, 34
Amino acids	5	2 to 6	25.1 ± 1.0	35, 36
N-acetyl-N-methylamino acid amides	8	5 to 9	25.1 ± 1.3	12
N-acetylamino acid amides	5	4 to 8	26.8 ± 0.4	14
2,5-diketopiperazines	4	4 to 6	26.7 ± 1.5	14
N-alkylureas (mono-, di-, tri-,				
and tetra-substituted)	20	1 to 9	27.3 ± 1.7	13, 37
N-alkylthioureas	7	1 to 5	26.8 ± 0.7	15
Li-alkanoates	7	1 to 7	26.3 ± 1.8	38-41
Na-alkanoates	5	1 to 16	24.4 ± 0.5	42-45
Tl-alkanoates	3	7 to 12	23.6 ± 0.4	46-48

TABLE 4. Heat capacity at T = 298.15 K of the CH₂ group contribution to $C_{p,m}$ of solid alkane- α , ω -diols compared with the values derived from various series of homologous alkyl compounds in the literature; N denotes the number of compounds in the series, and N(C) the number of C atoms in the compounds at the extremities of the series

chains can be differently packed and be allowed to give a progressively higher contribution to the total heat capacity. Therefore, in amino acids and their various derivatives, as well as in *N*-alkylureas and *N*-alkylthioureas, the increasing role of H-bond networks may account for the higher heat capacity contribution of the CH₂ group.

On the other hand, the increasing effect of the H-bond network can be observed by comparing the melting temperatures of compounds with the same N(C) belonging to different series, *e.g.* C₅H₁₂ (*n*-pentane), $T_{fus} = 143.4$ K; C₅H₁₂O (*n*-pentan-1-ol), $T_{fus} = 195.6$ K; C₅H₁₀O₂ (*n*-pentanoic acid), $T_{fus} = 239.2$ K; and C₅H₁₂N₂O₂ (*N*-acetyl-L-alaninamide), $T_{fus} = 436.4$ K; C₅H₁₀O (1,3-diethyl-2-urea), $T_{fus} = 386.5$ K; and C₅H₁₀S (1,3-diethyl-2-thiourea), $T_{fus} = 350.0$ K.

The three families of metal alkanoates, which show a progressive slight decrease of the contribution to $C_{p,m}$ of their CH₂ group as a function of the increasing size of the metal ion, should not be included in this interpretation scheme and require data from other series to be better understood.

In conclusion, the value of $(23.05 \pm 0.98) \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ derived in this work for the contribution of the CH₂ group to the $C_{p,\text{m}}$ of alkane- α , ω -diols is closer to the weighted mean of $(22.1 \pm 0.5) \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ for *n*-alkanes, *n*-alkanol-1-ols, and *n*-alkanoic acids than to the corresponding value of $(26.6 \pm 0.9) \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ for the *N*-containing substances.

Lastly, it is worth noting that the value of $21.92 \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$ for the heat capacity of the CH₂ group contribution in solid organic compounds recently suggested by Domalski and Hearing⁽⁴⁹⁾ on the basis of a group additivity scheme is decidedly closer to the weighted mean for *n*-alkanes, *n*-alkan-1-ols, and *n*-alkanoic acids than to that obtained from the other series of solid organic compounds reported in table 4, or even to the weighted mean for all the compounds reported therein, $(24.3 \pm 0.9) \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$.

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