Speeds of Sound and Excess Volumetric Properties of Mixtures of Water with 2-Propanol and with Ethylene Glycol Monoisopropyl Ether at 298.15 K

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Ultrasonic speeds of binary mixtures of water with 2-propanol and with ethylene glycol monoisopropyl ether have been measured at 298.15 K over the entire composition range. Density values were taken from the literature and combined with the measured values of the ultrasonic speeds to furnish estimates of the quantities $-(\partial V/\partial p)_s$, their excess counterparts, and the deviations of the ultrasonic speeds from the values calculated for ideal mixtures. The results have been analyzed using a segmented-composition model and compared with those previously obtained for related binary aqueous systems.

Introduction

Our interest lies in exploring the extent to which the composition dependence of a variety of excess thermodynamic properties of nonionic amphiphile + water systems provides evidence for the existence of ordered patterns of molecular aggregation. In earlier papers, we have reported the results of our measurements of some thermodynamic properties of the aqueous mixtures of the family of poly(ethylene glycol) monoalkyl ethers, whose members possess the generic formula $C_mH_{2m+1}(OC_2H_4)_nOH$ (C_mE_n) *(1-3)*. These investigations were concerned with the sensitivity to the alkyl chain length of the composition dependence of the concerned excess molar properties, together with the deviations of the ultrasonic speed from a quantity assumed to represent its value in an ideal mixture. Our intention is to extend the field of our investigations to other types of structural modifications, namely, the branching in the alkyl group for isomeric species and the changes in the polar head. Our attention has been drawn to the interesting aspects of the composition dependence of the excess molar volumes of the aqueous solutions of two couples of model isomeric species, 1-propanol *(4)* and 2-propanol *(5)* on the one hand, 2-propoxyethanol *(6)* and ethylene glycol monoisopropyl ether *(7)* on the other. Therefore, we thought it worthwhile to perform measurements of the ultrasonic speed of the 2-propanol + water and ethylene glycol monoisopropyl ether + water systems. There have been several investigations of ultrasonic speeds for the former system, but imprecision of measurements or sparseness of the results seemed to warrant a more detailed investigation of the mole fraction dependence of the compressibilities *(8, 9)*.

Experimental Section

Apparatus and Procedures. The speeds of sound, u, in both the pure liquids and their mixtures were measured at 298.15 K, using the sing-around technique which involves the passing of successive pulses of 1.8 MHz acoustic waves through the solution cell, with the arrival of one pulse triggering the transmission of the next *(10)*. The ultrasonic-pulse repetition rate (frequency f) was obtained by means of a sonic solution monitor (Mapco Inc., Nusonics Model 6105) and determined by a time-counter Hewlett-Packard 5384A. That rate is related to the speed of sound by a known explicit function of temperature and composition. The electronic processing circuit was coupled with a solution cell which is an 8 cm long brass cylinder (designed by E. Høgseth, Department of Chemistry, University of Bergen, Norway). The cell was submerged in an ultrathermostat, Heto 04 PE 623, which was provided with an auxiliary cooling device, Heto CB7. The temperature was controlled to within ±0.002 K and measured with a Hewlett-Packard calibrated quartz thermometer of type 2804A. Measurements of the frequency f were performed 20 min after the cell was dipped into the water bath and repeated 10 min later. Each sample measurement was bracketed by two water measurements, since water was used as a calibrant to determine the ultrasonic speed of the samples. It is appropriate to note that the errors in the measured ultrasonic speeds have been demonstrated to depend upon the difference between those values and that of the reference fluid *(11)*. The maximal error of the speeds measured relative to water [1496.687 $m\cdot s^{-1}$ at 298.15 K *(12)*] is believed to be less than 0.2 $m\cdot s^{-1}$; the reproducibility of the measurements is 4 times better.

Materials. Water was first deionized by means of ion-exchange resins and then doubly distilled. Its conductivity was always less than 1.0×10^{-6} $\Omega^{-1}\cdot cm^{-1}$. 2-Propanol (Aldrich HPLC grade) and ethylene glycol monoisopropyl ether (Tokyo Kasei Kogyo Co.) were used without further purification. The claimed purities for the two chemicals were, respectively, 99.5 and more than 99 mass %. All samples were used directly from the manufacturers' bottles, which were kept tightly sealed to minimize, as far as possible, the absorption of atmospheric water and carbon dioxide. The measured properties and those given in the literature are given in Table 1.

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Table 1. Comparison of Densities ϱ, Extensive Properties $A_p = -(\partial V_m/\partial T)_p$ (Equal to the Product of the Molar Volumes V_m and the Isobaric Thermal Expansivities α_p), Molar Isobaric Heat Capacities C_p,[n] and Speeds of Sound u of Liquids with the Literature at 298.15 K

	ϱ/(kg·m^{-3})		A_p/(mm^3·K^{-1}·mol^{-1})		C_p/(J·K^{-1}·mol^{-1})		u/(m·s^{-1})	
	exptl	lit.[a–f]	exptl	lit.[a,d,g,h]	exptl	lit.[a,i–k]	exptl	lit.[c,l,m]
water	–	997.048[a,n]	–	4.647[a,n]	–	75.292[a,n]	–	1496.687[l,n] 1497.00[c]
2-propanol	–	780.69[b,n] 780.62[c] 780.80[d]	–	83.79[d,n] 82.37[g] 81.91[h]	–	161.2[i,n] 154.55[j]	1137.07	1138.09[c] 1134.26[m]
2-ethylene glycol monoisopropyl ether	–	899.42[e,n] 899.6[f]	–	115.2[g,n]	–	238.8[k,n]	1266.27	–

[a] Reference 13. [b] Reference 14. [c] Reference 8. [d] Reference 5. [e] Reference 7. [f] Reference 15. [g] Reference 16. [h] Reference 17. [i] Reference 18. [j] Reference 19. [k] Reference 20. [l] Reference 12. [m] Reference 9. [n] Value chosen for the calculation of the quantities K_S, K_S^E, and u^D.

Mixtures were prepared by mass at room temperature with a precision of 0.1 mg from freshly thoroughly degassed samples of water and of organic component. They were stored in air-tight bottles. Corrections for buoyancy were included in the calculation of the mole fractions which are believed to be accurate to within 1 part in 10^4.

Results and Discussion

It proved to be impractical to make ultrasonic speed measurements at the same concentrations as those used previously for density measurements *(7, 14)*. Excess molar volumes V_m^E at the mole fractions of the ultrasonic speed measurements were obtained by cubic spline interpolation.

The molar quantity $K_S = V_m\kappa_S = -(\partial V_m/\partial p)_S$ has no universally acceptable "correct" name, since it is customary to reserve the term "compressibility" for the intensive property $\kappa_S = V_m^{-1}(\partial V_m/\partial p)_S$. κ_S and K_S are calculated from the Newton–Laplace equation

$$\kappa_S = (\varrho u^2)^{-1} = V_m(Mu^2)^{-1} \qquad (1)$$

$$K_S = V_m^{\,2}(Mu^2)^{-1} \qquad (2)$$

where ϱ is the density and M the molar mass of the mixture. Equations 1 and 2 are valid, provided that there is a negligible amount of absorption of the acoustic wave. In that case, the ultrasonic speed may be regarded as being a "true" thermodynamic quantity *(21)*.

Defining the ideal quantity K_S^{id} is not as straightforward as for the ideal molar volume V_m^{id} *(2)*. One may write a defining equation in terms of other well-established ideal quantities

$$K_S^{id} = K_T^{id} - T(A_p^{id})^2/C_p^{id} \qquad (3)$$

where $K_T = -(\partial V_m/\partial p)_T$, $A_p = (\partial V_m/\partial T)_p$, and C_p is the molar isobaric heat capacity *(22)*. The ideal values of all three of these quantities are the mole fraction weighted adducts of the corresponding properties of the pure components.

The corresponding excess molar quantity K_S^E is calculated from the equation

$$K_S^E = K_S - K_S^{id} \qquad (4)$$

Uncertainties in the values of A_p and C_p for the amphiphilic species lead to uncertainties on the order of 0.1–0.2 m^3·PPa^{-1}·mol^{-1} at the extrema of K_S^E. The level of internal consistency is an order of magnitude better.

Insofar as the Newton–Laplace equation is valid, the ultrasonic speed may be expressed solely in terms of thermodynamic quantities

$$u = V_m(MK_S)^{-1/2} \qquad (5)$$

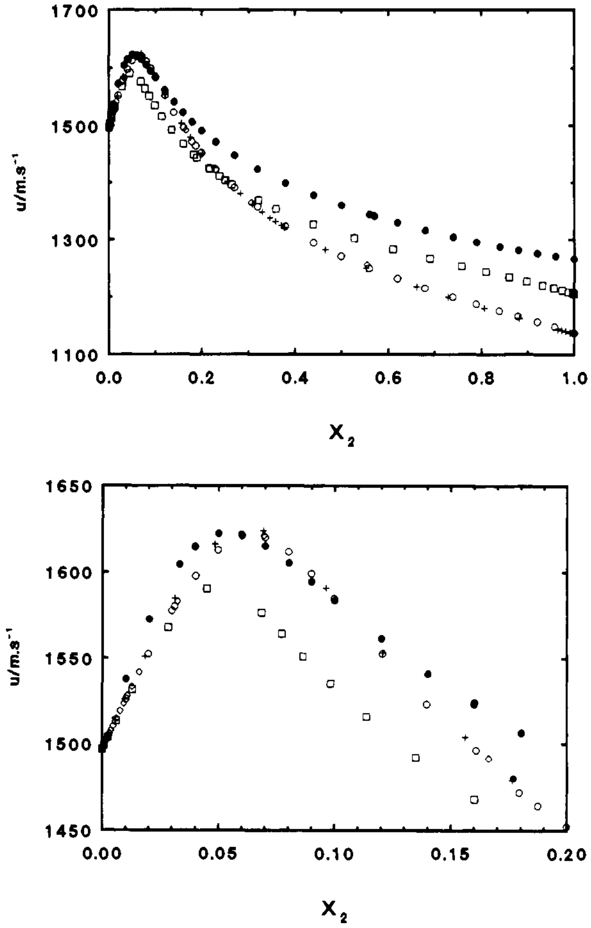

Figure 1. (a, top) Experimental ultrasonic speeds at 298.15 K for the binary mixtures containing $(1 - x_2)$ water (1) + x_2 1-propanol (2) [(□) ref 22], $(1 - x_2)$ water (1) + x_2 2-propanol (2) [(○) this work; (+) ref 9; (◇) ref 8], and $(1 - x_2)$ water (1) + x_2 ethylene glycol monoisopropyl ether (2) [(●) this work]. (b, bottom) Experimental ultrasonic speeds at 298.15 K in the water-rich region for the binary mixtures containing $(1 - x_2)$ water (1) + x_2 1-propanol (2) [(□) ref 22], $(1 - x_2)$ water (1) + x_2 2-propanol (2) [(○) this work; (+) ref 9; (◇) ref 8], and $(1 - x_2)$ water (1) + x_2 ethylene glycol monoisopropyl ether (2) [(●) this work].

This implies that we can calculate the ultrasonic speed u^{id} in an "ideal" mixture

$$u^{id} = V_m^{id}(MK_S^{id})^{-1/2} \qquad (6)$$

and its deviation u^D from the measured values for the same composition such as *(1)*

$$u^D = u - u^{id} \qquad (7)$$

The values of u, K_S, K_S^E, and u^D are reported in Table 2.

Table 2. Mole Fractions x_2, Extensive Properties $K_S = -(\partial V_m/\partial p)_S$, Excess Molar Quantities K_S^E, Deviations u^D from the Ideal Ultrasonic Speed of Water (1) with 2-Propanol (2) and with Ethylene Glycol Monoisopropyl Ether (2) at 298.15 K

x_2	u/ (m·s^{-1})	K_S^{a}/ (m^3·PPa^{-1}·mol^{-1})	K_S^E ^{a}/ (m^3·PPa^{-1}·mol^{-1})	u^D/ (m·s^{-1})
Water (1) + 2-Propanol (2)				
0	1496.687	8.09	0	0
0.010 23	1525.88	8.07	−0.82	67.7
0.019 91	1552.31	8.04	−1.60	125.0
0.030 04	1577.40	8.04	−2.38	177.9
0.031 18	1579.81	8.04	−2.47	183.2
0.039 99	1597.93	8.07	−3.12	222.0
0.049 76	1612.69	8.15	−3.78	257.0
0.060 19	1620.85	8.31	−4.42	284.3
0.070 05	1620.00	8.55	−4.92	299.4
0.080 06	1611.80	8.88	−5.34	305.6
0.089 93	1599.19	9.28	−5.69	305.7
0.099 84	1584.51	9.72	−5.99	302.5
0.120 46	1552.76	10.72	−6.52	291.4
0.139 69	1523.00	11.74	−6.92	277.5
0.160 94	1496.35	12.86	−7.35	265.6
0.179 35	1471.80	13.92	−7.63	251.8
0.187 49	1464.34	14.35	−7.79	248.6
0.199 85	1452.24	15.03	−8.00	242.5
0.229 93	1423.68	16.77	−8.41	226.4
0.270 08	1391.08	19.17	−8.86	206.9
0.319 69	1357.24	22.25	−9.24	185.0
0.379 64	1322.77	26.15	−9.48	160.9
0.439 83	1294.40	30.22	−9.51	139.9
0.499 53	1270.58	34.39	−9.36	121.3
0.559 26	1249.86	38.71	−9.02	104.4
0.619 17	1231.50	43.18	−8.51	88.8
0.679 10	1214.83	47.81	−7.82	74.1
0.738 97	1199.47	52.58	−6.94	60.1
0.788 84	1187.33	56.71	−6.04	48.8
0.838 76	1175.58	60.98	−4.98	37.7
0.878 69	1166.32	64.53	−4.00	28.8
0.920 43	1156.64	68.36	−2.84	19.4
0.957 28	1147.98	71.89	−1.66	10.8
0.998 33	1137.50	76.09	−0.07	0.4
1	1137.07	76.26	0	0
Water (1) + Ethylene Glycol Monoisopropyl Ether (2)				
0	1496.687	8.09	0	0
0.010 03	1537.49	8.05	−0.87	69.5
0.020 29	1572.51	8.06	−1.70	128.8
0.033 28	1604.43	8.19	−2.62	185.8
0.039 88	1614.65	8.32	−3.03	206.8
0.050 01	1622.29	8.58	−3.58	229.1
0.059 99	1621.48	8.95	−4.01	240.8
0.070 04	1615.03	9.39	−4.37	245.3
0.080 23	1605.28	9.88	−4.68	245.3
0.090 07	1594.49	10.40	−4.93	242.8
0.100 02	1583.41	10.95	−5.17	239.1
0.120 02	1561.46	12.08	−5.58	229.8
0.140 17	1541.10	13.28	−5.93	219.7
0.160 02	1522.96	14.49	−6.23	209.8
0.180 12	1506.28	15.75	−6.49	200.0
0.200 30	1491.15	17.03	−6.73	190.7
0.229 93	1471.26	18.95	−7.02	177.6
0.270 06	1447.99	21.63	−7.30	161.4
0.319 98	1423.54	25.06	−7.52	143.1
0.379 97	1398.95	29.32	−7.60	123.5
0.440 09	1378.02	33.74	−7.48	106.0
0.500 00	1359.75	38.31	−7.18	89.9
0.559 98	1343.65	43.04	−6.69	75.3
0.570 54	1340.97	43.88	−6.58	72.8
0.620 03	1329.20	47.90	−6.04	61.9
0.680 01	1316.33	52.83	−5.31	49.6
0.740 04	1304.40	57.85	−4.47	38.1
0.789 98	1295.62	62.08	−3.71	29.5
0.840 67	1287.55	66.45	−2.86	21.5
0.880 04	1281.72	69.85	−2.17	15.6
0.920 10	1276.17	73.32	−1.47	10.1
0.960 00	1271.01	76.79	−0.75	4.8
1	1266.27	80.29	0	0

^{a} 1 PPa = 10^{15} Pa.

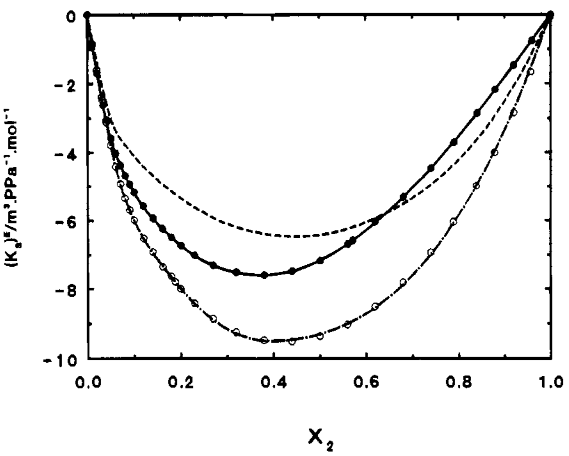

Figure 2. Excess molar quantities K_S^E at 298.15 K for the binary mixtures containing $(1 - x_2)$ water (1) + x_2 1-propanol (2) [(- - -) four-segment model equations applied to data from ref 22], $(1 - x_2)$ water (1) + x_2 2-propanol (2) [(○) this work; (· · ·) four-segment model equations applied to this work], and $(1 - x_2)$ water (1) + x_2 ethylene glycol monoisopropyl ether (2) [(●) this work; (—) four-segment equations applied to this work].

The values of the ultrasonic speeds are presented in Figure 1. Our results for the 2-propanol + water system agree closely in the water-rich region and for the pure solvent with the literature data *(8)*. Some differences are, however, observed in the amphiphile-rich region *(9)*. Figure 1 also shows the ultrasonic speeds of 1-PrOH + water mixtures *(23)*. The ultrasonic speeds for this system and the 2-PrOH + water and ethylene glycol monoisopropyl ether + water mixtures have similar mole fraction dependence profiles. The sharp increase in u with amphiphile content in water-rich mixtures is characteristic of all C_mE_n + water systems. The well-defined maximum tends to decrease with both increasing alkyl chain length and increasing polar head group size. It is unfortunate that we have been unable to obtain commercially synthesized 2-propoxyethanol.

The shapes of the K_S^E curves, as shown in Figure 2, are strongly reminiscent of the excess molar volumes. In both cases, the 2-PrOH + water system does not exhibit the same sharp change of slope at $x_2 = 0.06$ that is characteristic of 1-PrOH + water. Of some interest is the partial excess molar property K_S of water, defined as

$$K_{S,W}^E = K_S^E - \partial K_S^E/\partial x_A \tag{8}$$

where x_A holds for the mole fraction of the amphiphile A. The composition dependence of $K_{S,W}^E$ in the water-rich region is shown in Figure 3a. We see significant changes of slope for all three systems, with that for 1-ProH + water being more pronounced than for the aqueous mixtures of either of the branched species. At amphiphile-rich compositions, as seen in Figure 3b, both of the alkanol + water mixtures show rapid decreases which are clearly not evident for the ethylene glycol monoisopropyl ether + water mixtures (or any other alkoxyethanol + water system).

The changes of slope that have been observed for the excess and the partial excess molar properties of 1-PrOH + water have been observed to a more marked degree for the micelle-forming C_6E_n + water systems and the "borderline" surfactant C_4E_1 + water *(1, 3, 24)*. The rationale is that, at a critical micelle concentration or its analog, the amphiphile undergoes a significant change in its mode of self-aggregation. 1-Propanol appears to be able to form some type of labile micelle-like clusters more effectively than its isomer.

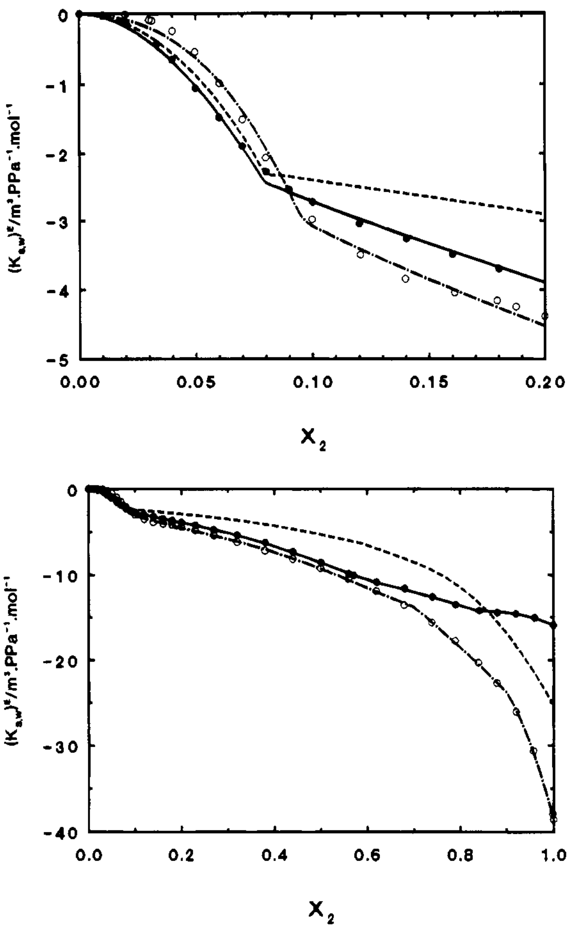

Figure 3. (a, top) Partial excess molar compressibilities $K^{\mathrm{E}}_{S,\mathrm{W}}$ of water at 298.15 K in the water-rich region for the binary mixtures containing $(1 - x_2)$ water (1) + x_2 1-propanol (2), $(1 - x_2)$ water (1) + x_2 2-propanol (2), and $(1 - x_2)$ water (1) + x_2 ethylene glycol monoisopropyl ether (2). Symbols have the same meaning as in Figure 2. (b, bottom) Partial excess molar compressibilities $K^{\mathrm{E}}_{S,\mathrm{W}}$ of water at 298.15 K for the binary mixtures containing $(1 - x_2)$ water (1) + x_2 1-propanol (2), $(1 - x_2)$ water (1) + x_2 2-propanol (2), $(1 - x_2)$ water (1) + x_2 ethylene glycol monoisopropyl ether (2). Symbols have the same meaning as in Figure 2.

Table 3. Optimized Four-Segment Model Parameters for the Excess Molar Quantities K^{E}_{S} of Water (1) with 1-Propanol (2), with 2-Propanol (2), and with Ethylene Glycol Monoisopropyl Ether (2) at 298.15 K[a]

	1-PrOH	2-PrOH	2-EGMIPE
x_{A}	0.060	0.075	0.052
x_{B}	0.225	0.275	0.225
x_{C}	0.725	0.650	0.550
a_{W}	−67.6	−79.7	−88.44
b_{W}	−38.0	−58.0	240
c_{W}	4105	2707	2533
q_{W}	−2.84	−4.60	−3.78
q_{A}	−1.02	−0.59	2.74
b_{L}	−17.9	−27.0	−25.9
a_{A}	−30.0	−41.0	−18.4
b_T	1.90	4.20	2.90
$10^2\sigma$	0.75	1.75	0.95

[a] Unit of K^{E}_{S} and coefficients: $\mathrm{m^3 \cdot PPa^{-1} \cdot mol^{-1}}$ (1 PPa = 10^{15} Pa).

The difference in the shapes of the amphiphile-rich regions of the partial excess molar properties of water for alkanols on the one hand and alkoxyethanols on the other must reflect important differences in the modes of self-aggregation of the pure amphiphiles. In the alkanol case, addition of water has a profound effect upon the presumed cyclic oligomeric structure. In the alkoxyethanol case, it appears that water is incorporated into the amphiphile structure without any significant change in the latter's scheme of self-aggregation.

The calculated values of the molar quantity K^{E}_{S} have been fitted by the most recent set of four-segment model equations *(3)*. These equations are given in the Appendix. This model, as its name implies, involves analyzing the excess molar property data of amphiphile + water systems on the assumption that the total composition range can be treated as being made up of four reasonably discrete regions (segments). The version of the model maintains continuity of $\mathrm{d}Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}$ at each of the three junction segments but derives its fitting capability from allowing discontinuities in $\mathrm{d}^2Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}^2$. The values of the optimized four-segment model parameters for the three 1-propanol +, 2-propanol +, and ethylene glycol monoisopropyl ether + water systems are summarized in Table 3.

Appendix. Four-Segment Model Equations

Water-Rich Segment.

$$0 \le x_{\mathrm{A}} \le x_1$$

$$Q^{\mathrm{E}} = a_{\mathrm{W}}(x_{\mathrm{A}} - x_{\mathrm{A}}^3) + b_{\mathrm{W}}(x_{\mathrm{A}}^2 - x_{\mathrm{A}}^3) + c_{\mathrm{W}}x_{\mathrm{A}}^3 \qquad \text{(a)}$$

where Q^{E} represents an excess molar property and x_{A} the mole fraction of the amphiphile A.

Pseudolamellar Segment.

$$x_2 \le x_{\mathrm{A}} \le x_3$$

$$Q^{\mathrm{E}} = q_{\mathrm{W}}(1 - x_{\mathrm{A}}) + q_{\mathrm{A}}x_{\mathrm{A}} + b_{\mathrm{L}}x_{\mathrm{A}}(1 - x_{\mathrm{A}}) \qquad \text{(b)}$$

Organic-Rich Segment.

$$x_3 \le x_{\mathrm{A}} \le 1$$

$$Q^{\mathrm{E}} = a_{\mathrm{A}}(1 - x_{\mathrm{A}})(1 - f_{\mathrm{a}})^2 + 3[Q^{\mathrm{E}}(x_3)](3f_{\mathrm{a}}^2 - 2f_{\mathrm{a}}^3) + (1 - x_{\mathrm{A}})(1 - f_{\mathrm{a}})^2 \qquad \text{(c)}$$

where

$$f_{\mathrm{a}} = (1 - x_{\mathrm{A}})/(1 - x_3)$$

$$[Q^{\mathrm{E}}(x_3)] = q_{\mathrm{W}}(1 - x_3) + q_{\mathrm{A}}x_3 + b_{\mathrm{L}}x_3(1 - x_3)$$

$$\{[\mathrm{d}Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}](x_3)\} = q_{\mathrm{A}} - q_{\mathrm{W}} + b_{\mathrm{L}}(1 - 2x_3)$$

Transitional Segment.

$$x_1 \le x_{\mathrm{A}} \le x_2$$

$$Q^{\mathrm{E}} = b_T(f_T^2 - 2f_T^3 + f_T^4) + [Q^{\mathrm{E}}(x_1)](4f_T^3 - 3f_T^4) + [Q^{\mathrm{E}}(x_2)](1 - 4f_T^3 + 3f_T^4) + \{[\mathrm{d}Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}](x_1)\}(x_2 - x_1) \times (f_T^3 - f_T^4) - \{[\mathrm{d}Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}](x_2)\}(x_2 - x_1)(f_T - f_T^3 - f_T^4) \qquad \text{(d)}$$

where

$$f_T = (x_2 - x_{\mathrm{A}})/(x_2 - x_1)$$

$$[Q^{\mathrm{E}}(x_1)] = a_{\mathrm{W}}(x_1 - x_1^3) + b_{\mathrm{W}}(x_1^2 - x_1^3) + c_{\mathrm{W}}x_1^3$$

$$\{[\mathrm{d}Q^{\mathrm{E}}/\mathrm{d}x_{\mathrm{A}}](x_1)\} = a_{\mathrm{W}}(1 - 3x_1^2) + b_{\mathrm{W}}(2x_1 - 3x_1^2) + 3c_{\mathrm{W}}x_1^2$$

$$[Q^E(x_2)] = q_W(1 - x_2) + q_A x_2 + b_L x_2(1 - x_2)$$

$$\{[dQ^E/dx_A](x_2)\} = q_A - q_W + b_L(1 - 2x_2)$$

a_w is the apparent excess molar property of the amphiphile A at infinite aqueous dilution. b_W and c_W are solute–solute interaction parameters. The former has been associated with hydration cosphere overlap and the latter with solute cluster formation. q_W and q_A are excess molar properties of water and the amphiphile, respectively, in a hypothetical pseudolamellar state. b_L is some type of interface property of that state. a_A is the apparent excess molar property of water at infinite dilution in the amphiphilic solute. b_T is a data-fitting parameter related to the range of compositions that are roughly correlated with miscibility gaps.

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