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Thermodynamic Properties of *n*-Alkoxyethanols + Organic Solvent Mixtures. XI. Total Vapor Pressure Measurements for *n*-Hexane, Cyclohexane or *n*-Heptane + 2-Ethoxyethanol at 303.15 and 323.15 K

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Total vapor pressures at 303.15 and 323.15 K were measured for binary systems of *n*-hexane, cyclohexane, or *n*-heptane + 2-ethoxyethanol. Measurements were made with a Van Ness type apparatus and were fitted to the modified Margules equation using Barker's method. The five-parameter modified Margules equation represents the measurements to within an average absolute deviation of approximately 0.01 kPa. The measurements reveal positive deviations from Raoult's law. Mixtures with *n*-heptane and cyclohexane show azeotropic behavior at both temperatures.

Introduction

The OH/O project is a part of a general program, the so-called TOM project,[1,2] in which mixtures containing the oxygen (O) and/or hydroxyl (OH) groups are investigated in order to characterize their interactions. Particular attention is paid to intra- and intermolecular effects, related to the presence of the O and/or OH groups in the same or different molecules.

We have contributed largely to this project by reporting experimental data at 298.15 K for excess enthalpies,[3,4] H^E, excess volumes,[5] V^E, and heat capacities at constant pressure,[5] C_p^E, of mixtures of alkoxyethanols (2-methoxyethanol, 2-ethoxyethanol, 2-butoxyethanol, 2-(2-methoxyethoxy)ethanol, 2-(2-ethoxyethoxy)ethanol, 2-(2-butoxyethoxy)ethanol) with organic solvents. We have also provided liquid−liquid equilibrium (LLE) measurements[6,7] for 2-methoxyethanol, 2-ethoxyethanol, 2-(2-methoxyethoxy)ethanol, or 2-(2-ethoxyethoxy)ethanol + alkane mixtures.

In continuation of this experimental work, we present vapor−liquid equilibrium VLE data for the mixtures *n*-hexane, cyclohexane, or *n*-heptane + 2-ethoxyethanol at 303.15 and 323.15 K. These results extend the database available for a class of mixtures which is very interesting from a theoretical point of view due to the strong intramolecular effects related to the presence of the OH and O groups in the same molecule (alkoxyethanols).[8] In the framework of the TOM project, our final goal is the characterization of these mixtures in terms of the DISQUAC group contribution model.[1,2]

Experimental Section

Materials. All chemicals were obtained from Fluka and had percent purities (by chromatographic analysis, as given by the manufacturer for each specific lot) of 99.6% (*n*-hexane), 99.9% (cyclohexane), 99.9% (*n*-heptane), and 99.9% (2-ethoxyethanol). All chemicals were degassed by vacuum distillation and were used without additional purification. The pure component vapor pressures measured in this study are reported in Table 1 where they are compared with smoothed values obtained from the literature. Generally, good agreement is found between the reported results and those of the present study.

Table 1. Comparison of Pure Component Vapor Pressures P_i^{sat}/kPa

	303.15 K		323.15 K	
	exptl	lit.	exptl	lit.
n-hexane	24.938	24.969[a]	53.982	54.065[a]
cyclohexane	16.230	16.228[b]	36.180	36.230[b]
n-heptane	7.811	7.788[c]	18.869	18.899[c]
2-ethoxyethanol	1.015	0.971[d]	3.228	3.121[d]

[a] American Petroleum Institute Project 44.[9] [b]Willingham et al.[10] [c]TRC Thermodynamic Tables.[11] [d]Riddick et al.[12] and Antosik et al.[13]

Apparatus and Procedure. The apparatus is essentially the same as described in detail by Bhethanabotla and Campbell.[14] It is of the Van Ness type[15] in which total pressure is measured as a function of overall composition in the equilibrium cell. Two modifications to the apparatus described by Bhethanabotla and Campbell have been made: the pressure gauge has been replaced with one of 0.001 kPa resolution as described by Pradhan et al.[16] and the piston injectors have been replaced with Ruska pumps (model 2200) having a resolution of 0.001 cm^3.

The overall composition in the equilibrium cell is changed by charging metered amounts of the pure components from their respective piston injectors. The pressure in the cell is read after equilibration. The small correction (approximately 0.001 in mole fraction) to convert the overall mole fraction in the equilibrium cell to the liquid-phase mole fraction is made as part of the data reduction procedure as described by Bhethanabotla and Campbell.

Experimental uncertainties are ±0.1% in pressure, ±0.02 K in temperature, and between ±0.0005 and ±0.001

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Table 2. Total Pressure P as a Function of Liquid-Phase Mole Fraction x_1 for Alkane (1) + 2-Ethoxyethanol (2), at 303.15 and 323.15 K, Calculated Vapor-Phase Mole Fraction, y_1, and Natural Logarithm of Activity Coefficients

303.15 K					323.15K				
x_1	P/kPa	y_1	ln γ_1	ln γ_2	x_1	P/kPa	y_1	ln γ_1	ln γ_2
n-Hexane (1) + 2-Ethoxyethanol (2)									
0.0000	1.009	0.0000	2.1303	0.0000	0.0000	3.225	0.0000	2.0128	0.0000
0.0284	6.072	0.8378	1.9728	0.0058	0.0203	10.431	0.6948	1.8869	0.0054
0.0577	9.992	0.9035	1.8367	0.0143	0.0423	16.852	0.8145	1.7928	0.0113
0.0974	13.811	0.9320	1.6677	0.0308	0.0718	23.679	0.8707	1.6720	0.0223
0.1473	16.995	0.9463	1.4763	0.0586	0.1103	30.356	0.9016	1.5254	0.0403
0.1973	19.052	0.9532	1.3056	0.0959	0.1506	35.385	0.9175	1.3848	0.0638
0.2475	20.413	0.9572	1.1522	0.1402	0.1925	39.139	0.9270	1.2506	0.0929
0.3480	21.965	0.9614	0.8896	0.2541	0.2356	41.955	0.9331	1.1246	0.1300
0.3982	22.427	0.9627	0.7769	0.3206	0.2801	44.115	0.9375	1.0057	0.1714
0.3986	22.429	0.9627	0.7760	0.3214	0.3261	45.738	0.9407	0.8935	0.2213
0.4484	22.768	0.9637	0.6743	0.3957	0.3734	46.995	0.9431	0.7880	0.2802
0.4485	22.775	0.9637	0.6741	0.3958	0.3973	47.535	0.9442	0.7383	0.3107
0.4985	23.038	0.9645	0.5807	0.4801	0.4221	48.002	0.9451	0.6888	0.3466
0.5483	23.258	0.9652	0.4955	0.5740	0.4470	48.456	0.9460	0.6414	0.3831
0.5981	23.448	0.9658	0.4170	0.6813	0.4969	49.177	0.9476	0.5524	0.4627
0.6484	23.612	0.9665	0.3442	0.8016	0.5468	49.799	0.9490	0.4707	0.5526
0.6985	23.772	0.9673	0.2776	0.9380	0.5969	50.312	0.9503	0.3951	0.6546
0.7489	23.929	0.9681	0.2157	1.1031	0.6973	51.279	0.9530	0.2610	0.9037
0.7990	24.092	0.9692	0.1592	1.2978	0.7478	51.714	0.9546	0.2013	1.0601
0.8492	24.271	0.9707	0.1074	1.5427	0.7984	52.168	0.9566	0.1464	1.2475
0.8994	24.491	0.9730	0.0604	1.8739	0.8507	52.654	0.9592	0.0946	1.4950
0.9439	24.712	0.9771	0.0248	2.3016	0.8943	53.076	0.9626	0.0563	1.7614
0.9590	24.795	0.9797	0.0148	2.4980	0.9401	53.543	0.9692	0.0225	2.1446
1.0000	24.938	1.0000	0.0000	3.2750	0.9703	53.803	0.9785	0.0067	2.4929
					1.0000	53.982	1.0000	0.0000	2.9437
Cyclohexane (1) + 2-Ethoxyethanol (2)									
0.0000	1.023	0.0000	1.8166	0.0000	0.0000	3.230	0.0000	1.6971	0.0000
0.0294	3.601	0.7245	1.7011	0.0014	0.0282	8.160	0.6130	1.5887	0.0049
0.0590	5.719	0.8306	1.6024	0.0072	0.0574	12.416	0.7518	1.5018	0.0110
0.0987	7.964	0.8819	1.4779	0.0198	0.0973	17.069	0.8253	1.3871	0.0229
0.1483	10.043	0.9095	1.3329	0.0415	0.1467	21.481	0.8661	1.2548	0.0432
0.1982	11.546	0.9238	1.1983	0.0697	0.1966	24.802	0.8877	1.1300	0.0709
0.2484	12.640	0.9323	1.0725	0.1069	0.2470	27.304	0.9010	1.0125	0.1055
0.2985	13.439	0.9379	0.9565	0.1512	0.2971	29.172	0.9098	0.9039	0.1475
0.3487	14.031	0.9418	0.8485	0.2039	0.3473	30.606	0.9161	0.8025	0.1971
0.3989	14.474	0.9447	0.7481	0.2641	0.3977	31.722	0.9209	0.7079	0.2542
0.4491	14.812	0.9469	0.6549	0.3337	0.4482	32.572	0.9247	0.6196	0.3197
0.4992	15.090	0.9487	0.5680	0.4119	0.4636	32.832	0.9258	0.5940	0.3403
0.4993	15.068	0.9487	0.5682	0.4117	0.4985	33.286	0.9279	0.5379	0.3930
0.5490	15.271	0.9502	0.4880	0.5001	0.5484	33.851	0.9306	0.4623	0.4765
0.5498	15.297	0.9502	0.4867	0.5021	0.5488	33.857	0.9306	0.4617	0.4776
0.5980	15.425	0.9515	0.4145	0.5993	0.5980	34.311	0.9330	0.3921	0.5716
0.6492	15.597	0.9528	0.3432	0.7179	0.6102	34.422	0.9336	0.3757	0.5965
0.6992	15.722	0.9541	0.2787	0.8520	0.6983	35.077	0.9376	0.2641	0.8096
0.7493	15.840	0.9556	0.2189	1.0088	0.7487	35.408	0.9399	0.2058	0.9638
0.7997	15.951	0.9573	0.1631	1.2017	0.7988	35.706	0.9425	0.1517	1.1498
0.8494	16.068	0.9594	0.1122	1.4437	0.8491	35.966	0.9457	0.1013	1.3875
0.8996	16.177	0.9624	0.0647	1.7792	0.8997	36.238	0.9504	0.0553	1.7123
0.9398	16.282	0.9667	0.0307	2.1745	0.9396	36.359	0.9576	0.0247	2.0679
0.9700	16.338	0.9740	0.0100	2.6269	0.9705	36.401	0.9698	0.0072	2.4474
1.0000	16.230	1.0000	0.0000	3.3310	1.0000	36.180	1.0000	0.0000	2.9317
n-Heptane (1) + 2-Ethoxyethanol (2)									
0.0000	1.013	0.0000	2.3045	0.0000	0.0000	3.230	0.0000	2.1651	0.0000
0.0297	2.906	0.6609	2.1154	0.0043	0.0289	7.161	0.5599	1.9950	0.0047
0.0592	4.227	0.7717	1.9536	0.0125	0.0582	10.053	0.6932	1.8469	0.0129
0.0991	5.418	0.8264	1.7550	0.0300	0.0979	12.772	0.7651	1.6660	0.0294
0.1490	6.332	0.8551	1.5366	0.0615	0.1477	14.950	0.8049	1.4635	0.0585
0.1991	6.876	0.8693	1.3462	0.1021	0.2375	17.065	0.8353	1.1573	0.1322
0.2491	7.213	0.8775	1.1800	0.1502	0.2681	17.493	0.8409	1.0675	0.1633
0.2991	7.434	0.8827	1.0331	0.2059	0.2982	17.833	0.8454	0.9854	0.1956
0.3493	7.586	0.8864	0.9023	0.2683	0.3486	18.259	0.8512	0.8600	0.2558
0.3873	7.669	0.8885	0.8122	0.3207	0.3491	18.285	0.8512	0.8588	0.2568
0.3496	7.590	0.8864	0.9014	0.2687	0.3978	18.570	0.8556	0.7503	0.3214
0.3995	7.694	0.8890	0.7848	0.3393	0.3989	18.590	0.8557	0.7479	0.3229
0.4503	7.773	0.8912	0.6776	0.4177	0.4289	18.741	0.8580	0.6865	0.3664
0.4995	7.832	0.8929	0.5834	0.5033	0.4636	18.895	0.8604	0.6199	0.4204
0.5484	7.881	0.8945	0.4979	0.5972	0.5489	19.187	0.8657	0.4733	0.5711
0.5995	7.928	0.8961	0.4162	0.7076	0.5991	19.329	0.8685	0.3963	0.6753

Table 2. (Continued)

303.15 K					323.15K				
x_1	P/kPa	y_1	ln γ_1	ln γ_2	x_1	P/kPa	y_1	ln γ_1	ln γ_2
				n-Heptane (1) + 2-Ethoxyethanol (2)					
0.6494	7.966	0.8978	0.3433	0.8293	0.6490	19.466	0.8713	0.3257	0.7928
0.6995	8.004	0.8998	0.2761	0.9686	0.6991	19.569	0.8742	0.2598	0.9293
0.7496	8.041	0.9024	0.2149	1.1298	0.7492	19.673	0.8774	0.1987	1.0901
0.7997	8.080	0.9058	0.1591	1.3227	0.7996	19.751	0.8813	0.1419	1.2859
0.8495	8.115	0.9105	0.1085	1.5621	0.8498	19.822	0.8871	0.0909	1.5275
0.8999	8.149	0.9177	0.0622	1.8894	0.9001	19.829	0.8979	0.0471	1.8364
0.9394	8.159	0.9277	0.0301	2.2618	0.9399	19.732	0.9156	0.0199	2.1507
0.9693	8.108	0.9439	0.0104	2.6824	0.9717	19.479	0.9449	0.0054	2.4647
1.0000	7.811	1.0000	0.0000	3.3848	1.0000	18.869	1.0000	0.0000	2.8062

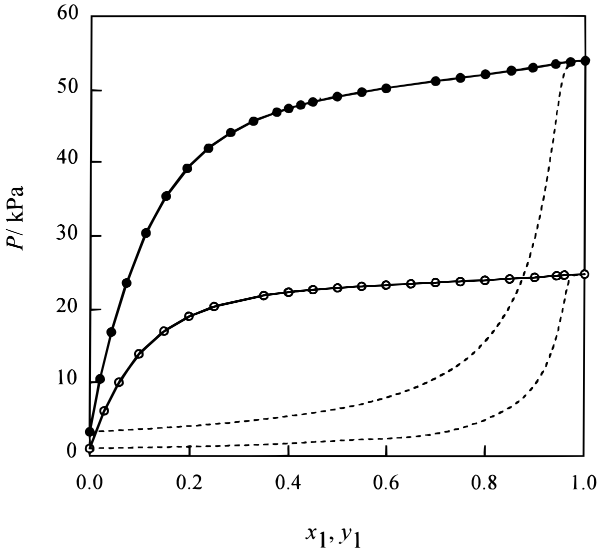

Figure 1. Pressure P versus liquid-phase mole fraction x_1 for *n*-hexane (1) + 2-ethoxyethanol (2): ○, experimental $P-x_1$ result at 303.15 K; ●, experimental $P-x_1$ result at 323.15 K; solid lines are fitted $P-x_1$ result using coefficients from Table 4; dashed lines are calculated $P-y_1$ results.

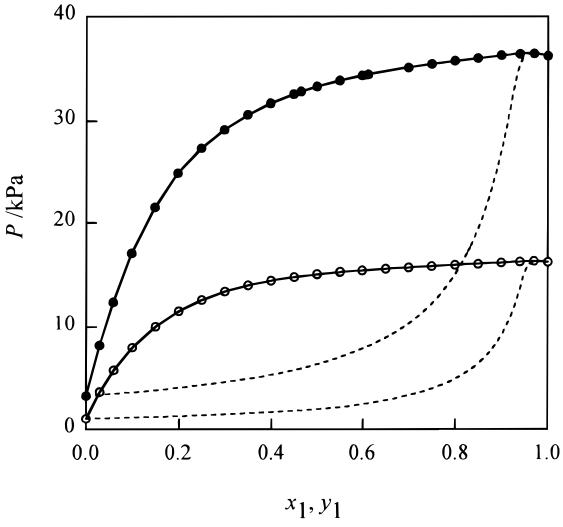

Figure 2. Pressure P versus liquid-phase mole fraction x_1 for cyclohexane (1) + 2-ethoxyethanol (2): ○, experimental $P-x_1$ result at 303.15 K; ●, experimental $P-x_1$ result at 323.15 K; solid lines are fitted $P-x_1$ result using coefficients from Table 4; dashed lines are calculated $P-y_1$ results.

in mole fraction, the smaller value applying at the extremes in composition.

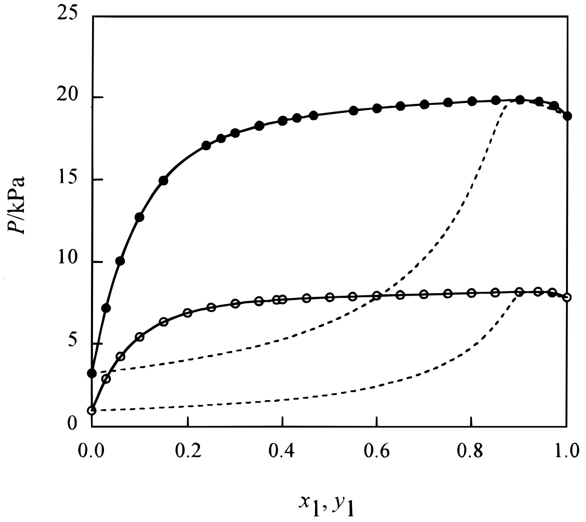

Figure 3. Pressure P versus liquid-phase mole fraction x_1 for *n*-heptane (1) + 2-ethoxyethanol (2): ○, experimental $P-x_1$ result at 303.15 K; ●, experimental $P-x_1$ result at 323.15 K; solid lines are fitted $P-x_1$ result using coefficients from Table 4; dashed lines are calculated $P-y_1$ results.

Table 3. Saturated Liquid Volumes V_i^{L} and Second Virial Coefficients for Single Components B_{ii} and Mixtures B_{ij} Used for Alkane (1) + 2-Ethoxyethanol (2) Systems[a] at 303.15 and 323.15 K

	B_{11}/cm^3·mol^{-1}	B_{12}/cm^3·mol^{-1}	V_i^{L}/cm·mol$^{3-1}$
	At 303.15 K		
n-hexane (1)	−1802	−1534	132.6
cyclohexane (1)	−1815	−1526	109.5
n-heptane (1)	−2806	−1893	148.4
	At 323.15 K		
n-hexane (1)	−1501	−1287	136.5
cyclohexane (1)	−1500	−1276	112.3
n-heptane (1)	−2285	−1571	152.4

[a] For 2-ethoxyethanol (2), B_{22}/cm^3·mol^{-1} = −3584 and V_2^{L}/cm^3·mol^{-1} = 97.8 at 303.15 K and B_{22}/cm^3·mol^{-1} = −2540 and V_2^{L}/cm^3·mol^{-1} = 99.8 at 323.15 K.

Experimental Results and Analysis

The total pressure P measured as a function of liquid-phase mole fraction, x_1, for the alkane (1) + 2-ethoxyethanol (2) mixtures considered are listed in Table 2. They are also plotted in Figures 1 to 3. No measurements have been found in the literature that may be compared directly to these. In fact, the only data we were able to locate for any of the systems studied here were the isobaric data (at 101 kPa) reported by Suryanarayana and Van Winkle[17] for *n*-hexane + 2-ethoxyethanol.

Because the experimental technique does not provide experimental data for y_1, the mole fraction in vapor phase

Table 4. Values of Parameters Appearing in Eq 1 and Resulting Average Deviation ΔP_{avg} and Maximum Deviations ΔP_{max} for Alkane (1) + 2-Ethoxyethanol (2) Systems

	A_{12}	A_{21}	α_{12}	α_{21}	η	ΔP_{avg}/kPa	ΔP_{max}/kPa
			At 303.15 K				
n-hexane (1)	2.1303	3.2750	1.6474	8.5556	3.0116	0.007	0.018
cyclohexane (1)	1.8166	3.3310	1.5299	10.5611	1.7019	0.007	0.018
n-heptane (1)	2.3045	3.3848	1.9899	10.7188	6.0369	0.002	0.008
			At 323.15 K				
n-hexane (1)	2.0128	2.9437	1.3616	5.5953	1.8439	0.013	0.085
cyclohexane (1)	1.6971	2.9317	1.1837	5.8514	0.4675	0.012	0.068
n-heptane (1)	2.1651	2.8062	1.3517	3.3287	1.1710	0.009	0.030

of component 1, the $P-x$ measurements were reduced using Barker's method[18] to obtain values of y_1 and γ_i, the activity coefficient of component i in liquid state. To this end, it was assumed that the latter are represented by the five-parameter form of the modified Margules equation[19]

$$\frac{G^E}{RT} = \sum x_i \ln \gamma_i = x_1x_2\left(A_{21}x_1 + A_{12}x_2 - \frac{\alpha_{12}\alpha_{21}x_1x_2}{\alpha_{12}x_1 + \alpha_{21}x_2 + \eta x_1x_2}\right) \quad (1)$$

The parameters in eq 1 were obtained by minimizing the sum of the squares between the measured and calculated pressures. Calculated pressures are obtained from

$$P_{calc} = x_1\gamma_1P_1^0 \exp\left\{\frac{-(B_{11} - V_1^L)(P - P_1^0) - P\delta_{12}y_2^2}{RT}\right\} + x_2\gamma_2P_2^0 \exp\left\{\frac{-(B_{22} - V_2^L)(P - P_2^0) - P\delta_{12}y_1^2}{RT}\right\} \quad (2)$$

where P_i^0 is the vapor pressure and V_i^L is the saturated liquid volume of pure component i at temperature T. The second virial coefficients are denoted by B_{ij} and $\delta_{12} = 2B_{12} - B_{11} - B_{22}$.

Equation 2 is valid assuming that the vapor phase of the mixture, as well as the vapor in equilibrium with the pure components, is described by the volume-explicit virial equation terminated after the second virial coefficient, V_i^L is constant over the pressure range considered, the liquid partial molar volume of each component is invariant with composition, and the standard states for γ_i are the pure components at the same T and P as those of the mixtures.

Values of second virial coefficients and saturated liquid volumes used in the calculations are given in Table 3. Second virial coefficients were determined from the Hayden−O'Connell[20] method. The constants needed for alkanes were taken from Fredenslund et al.[21] For 2-ethoxyethanol, the critical values of pressure and temperature are 4.180 MPa and 570 K, mean radius of gyration[22] = 3.31 Å, dipole moment[12] = 2.08 D, and association parameter[22] = 1.55. All solvation parameters were neglected.

Liquid volumes for 2-ethoxyethanol were obtained by smoothing densities reported by Riddick et al.[12] and Venkatesulu et al.[23] Values for *n*-hexane and *n*-heptane were taken from API Project 44[9] and for cyclohexane from Egloff.[24]

Parameters of the G^E model and resulting average and maximum deviations between calculated and experimental pressures are given for each system in Table 4. The data are represented by the G^E model generally to within an average of ±0.01 kPa with a maximum deviation of ±0.09 kPa. Calculated y_1 values are listed in Table 2 together with $\ln \gamma_i$ (i = 1, 2). Note that all investigated mixtures show positive deviation from Raoult's law. Azeotropic behavior is found for systems with cyclohexane and heptane at both 303.15 and 323.15 K. Coordinates of azeotropes are given in Table 5. The *n*-heptane (1) + 2-ethoxyethanol (2) system also presents an azeotrope[25] at atmospheric pressure, 369.65 K and $x_1 = 0.8468$.

Table 5. Calculated Azeotrope Coordinates for Alkane (1) + 2-Ethoxyethanol (2) Mixtures

alkane	T_{az}/K	P_{az}/kPa	$x_{1,az}$
cyclohexane	303.15	16.26	0.9837
	323.15	36.38	0.9706
n-heptane	303.15	8.13	0.9160
	323.15	19.82	0.8953

Conclusions

$P-x$ data have been measured at 303.15 and 323.15 K for mixtures of 2-ethoxyethanol with *n*-hexane, cyclohexane, or *n*-heptane. All systems show positive deviations from Raoult's law. Those with cyclohexane or *n*-heptane present azeotropic behavior.

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