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Studies of Monohydric Alcohols in Aqueous Sodium Dodecyl Sulfate Solutions at T = (298.15 and 308.15) K

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Densities, ρ, speeds of sound, u, viscosities, η, and specific conductivities, κ, of mixtures of 1-pentanol or 1-hexanol with the anionic surfactant sodium dodecyl sulfate (SDS) have been measured at molalities between (0.05 and 0.10) $\mathrm{mol \cdot kg^{-1}}$ and temperatures of (298.15 and 308.15) K. The apparent molar volumes, Φv, partial molar volumes, $\Phi^{o}v$, transfer volume, $\Phi^{o}v(\mathrm{tr})$, isentropic compressibility, κ_s, relative viscosity, η_{rel}, viscosity A- and B-coefficients, free energies per mole of solvent, $\Delta\mu_1^{0\#}$, and per mole of solute, $\Delta\mu_2^{0\#}$, and limiting molar conductivity, Λ^0_m, were calculated to permit speculation of the interactions prevailing in the studied aqueous surfactant−alcohol systems. The results were interpreted from the point of view of alcohol−surfactant−water interactions in these systems. Further, ^{1}H NMR chemical shift measurements in micelle solutions containing 1-pentanol have also been carried out. The results obtained are compared with the conclusions drawn from the thermodynamic measurements.

Introduction

Numerous studies of alcohol solubilization in the micelles of anionic surfactants, have been carried out utilizing different techniques with the aim of determining the decrease in the critical micelle concentration (cmc),[1,2] NMR techniques,[3−5] fluorescence,[6,7] thermodynamic measurements,[8−12] and various other methods.[13−17] The addition of alcohol can strongly influence the behavior of the micelles and increase or decrease the micellar size, depending on the hydrophilic/hydrophobic character of the alcohols.[18] The small chain or hydrophilic alcohols mainly solubilize in the aqueous solution and affect the micellization process by modifying the solvent properties. However, the medium chain or hydrophobic alcohol molecules take part in the micellization process and become unique components of the micelle aggregates.[17] The water, surfactant, and alcohol system is quite complex and is examined critically.[19] When the alcohol partitions between the water and the micellar pseudo phase, the fraction bound to the micelles replaces water molecules at the interface leading to increased electrostatic repulsion between surfactant head groups.[20,4] The moderately hydrophobic alcohols in low concentration promote micellization probably by residing at the micellar surface and reducing unfavorable water hydrocarbon contacts.[7] However, at higher concentrations these alcohols destabilize micelles by displacing water from the surface, therefore decreasing its effective dielectric constant, increasing headgroup repulsions, and disrupting surfactant packing.

In continuation of my previous work, the effect of alcohols on the micellization of the anionic surfactant sodium dodecyl benzene sulfonate (SDBS),[21] this paper presents the critical study of the micellization of aqueous sodium dodecyl sulfate, SDS, with monohydric alcohols, 1-pentanol and 1-hexanol. DeLisi et al.[11] and Hoiland et al.[12] have also investigated the partial molar volumes of the similar systems at T = 298.15 K. In this paper an attempt has been made to examine the micellar behavior of SDS in presence of medium chain alcohols using volumetric, viscometric, and conductometric techniques. The study shall be confined to the aqueous micellar phase by the assessment of apparent molar volume, Φv, partial molar volume $\Phi^{o}v$, transfer volume $\Phi^{o}v(\mathrm{tr})$, isentropic compressibility, κ_s, viscosity A- and B-coefficients, and limiting molar conductivity, Λ^0_m, using the experimental data of density, ρ, speed of sound, u, viscosity, η, and specific conductivities, κ, of the alcohols, 1-pentanol at molalities between (0.05 and 0.25) $\mathrm{mol \cdot kg^{-1}}$ and 1-hexanol at molalities between (0.01 and 0.08) $\mathrm{mol \cdot kg^{-1}}$ in (0.05 and 0.10) $\mathrm{mol \cdot kg^{-1}}$ aqueous SDS solutions at temperatures between (298.15 and 308.15) K. Apart from these, ^{1}H NMR studies have also been carried out for the present SDS−alcohol systems, the results of which are interpreted in terms of the approximate location of the alcohols in the micellar aggregates. An elaborate and critical study of the micellization and interfacial properties of SDS and its interaction with n-alcohols has been made. Further, the results obtained from the present investigations are compared with those in the case of SDBS with 1-pentanol or 1-hexanol, reported previously.[21]

Experimental Section

Materials. SDS was obtained from HiMedia Laboratories Pvt. Ltd., Mumbai, India, and was purified by the method given in literature.[22] The analytical grade 1-pentanol with a mass fraction purity of 0.99 and 1-hexanol with a mass fraction purity of 0.98 were purchased from Spectrochem Pvt. Ltd. Mumbai, India, and Acros Organics, New Jersey, respectively, and were purified by fractional distillation before use. Alcohols were kept over molecular sieves (0.4 nm Sigma Union Carbide type). The solubility of 1-pentanol and 1-hexanol in water is low,[21] and it increases in SDS micelle solution with the increase in surfactant concentration and temperature. However, it was assumed that the solubility of these alcohols in water would not cause a significant change in the cmc of SDS in water. Deuterium oxide (Aldrich, with an isotopic mass fraction purity of 0.999) was used without further purification as the solvent in ^{1}H NMR studies. Deionized, double-distilled water of conductance $1 \cdot 10^{-6}$ $\mathrm{S \cdot cm^{-1}}$ at 298.15 K was used for all measurements. Solutions

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Table 1. Speed of Sound, *u*, of Aqueous SDS Solutions at T = (298.15, 303.15, 308.15, and 313.15) K

m	u/m·s^{-1}			
mol·kg^{-1}	298.15 K	303.15 K	308.15 K	313.15 K
0.001	1497.2	1509.5	1520.1	1529.0
0.003	1497.5	1509.8	1520.4	1529.3
0.005	1497.8	1510.1	1520.6	1529.5
0.007	1497.9	1510.1	1520.7	1529.5
0.008	1497.9	1510.2	1520.7	1529.6
0.010	1498.1	1510.3	1520.8	1529.7
0.020	1498.2	1510.3	1520.8	1529.5
0.030	1498.4	1510.4	1520.7	1529.4
0.050	1498.3	1510.2	1520.3	1528.8
0.070	1498.2	1509.9	1519.9	1528.2
0.103	1498.2	1509.7	1519.3	1527.3

were prepared by weighing appropriate amount of the SDS in an electronic balance, (Afcoset-ER120A) with a precision of 0.0001 g. Double-distilled water and alcohols were degassed by vacuum pump shortly before sample preparation.

Apparatus and Procedure. The densities and speeds of sound of aqueous solutions of SDS and that with alcohols were simultaneously and automatically measured using a digital vibrating tube densimeter and speed of sound analyzer (Anton Paar DSA-5000) provided with automatic viscosity correction and two integrated Pt 100 thermometers. Both the speed of sound and the density are extremely sensitive to temperature, so it was kept constant within ± 0.001 K using a proportional temperature controller. The apparatus was first calibrated with triple-distilled water and dry air. Standard uncertainties in density measurements were estimated to be $\pm\ 2\cdot10^{-3}$ kg·m^{-3} and for the speed of sound are ± 0.1 m·s^{-1}. Further information about the experimental techniques has been given elsewhere.[23] The experimental values of the densities of aqueous SDS solutions at different temperatures are compared with the literature values,[9] and a good agreement is found. The literature values of the densities of aqueous SDS at different concentrations and temperatures are added in Table 1.

The kinematic viscosities of the pure liquids and their mixtures were measured at temperatures of (298.15 and 308.15) K and atmospheric pressure using Ubbelohde suspended level viscometer. Experimental details have been given previously.[23,24] The viscometer was filled with liquid or liquid mixtures, and its limbs were closed with Teflon caps taking due precaution to reduce evaporation losses. An electronic digital stopwatch with a readability of 0.01 s was used for flow time measurements. The measured values of kinematic viscosity, ν, were converted to dynamic viscosity, η, after multiplication by the density. The reproducibility of dynamic viscosity was found to be within ± 0.003 mPa·s. A thermostatically controlled, well-stirred water bath whose temperature was controlled to ± 0.01 K was used for all of the measurements.

Conductivities were measured using a digital conductivity meter (306) of Systronics which is a microcontroller-based instrument for measuring the specific conductivity of solutions using a conductivity cell. The conductivity cell was calibrated with standard KCl solution, and the obtained cell constant was 1.0 cm^{-1}. Conductivity measurements were carried out in a double-walled jacket containing solution in which water was circulated from a thermostat, and the temperature was maintained within ± 0.01 °C. Before starting the experiment, the system was equilibrated at the particular temperature for at least 30 min. The uncertainty of the specific conductivity measurements is ± 0.1 mS·cm^{-1}. The cmc of SDS in an aqueous solution was taken as the SDS concentration at the break point in the plot of specific conductance versus SDS concentration. The cmc of SDS in water was determined to be 8.2 mmol·dm^{-3} and was found to be in good agreement with the value reported in literature,[25] whereas at T = 308.15 K it was 8.7 mmol·dm^{-3} and agreed well with literature.[26]

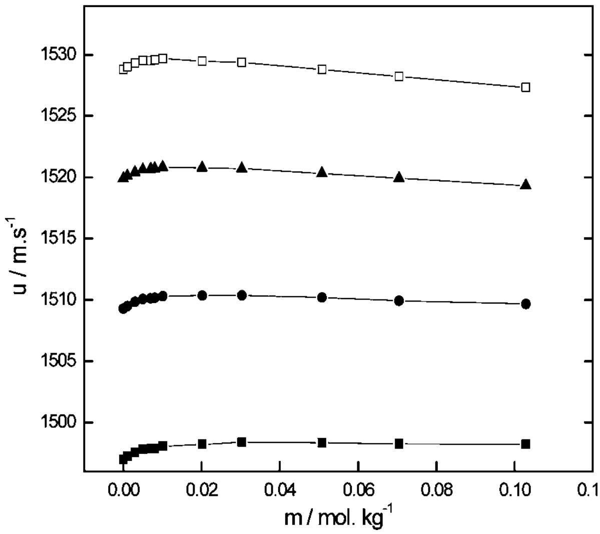

Figure 1. Speed of sound, *u*, in aqueous solutions of SDS as a function of molality: ■, 298.15 K; ●, 303.15 K; ▲, 308.15 K; and □, 313.15 K.

To investigate the effect of alcohols on the micellar phase, ^{1}H NMR measurements were performed on an Avance II 400 NMR spectrometer at a frequency of 400.13 MHz. Deuterium oxide was used as the solvent instead of water to weaken the water signal for all solutions. The method depends on the ability of the alcohols to affect the chemical shift of different proton signals of the surfactant molecules. These chemical shifts were measured in the presence of 1-pentanol as a function of both alcohol and surfactant concentration. The chemical shift measurements of various resonance peaks of SDS are given on the δ scale in parts per million (ppm) of the applied frequency.

Results and Discussion

The concentration dependence of speeds of sound of aqueous solution of SDS in the molality from (0.00 to 0.12) mol·kg^{-1} at T = (298.15, 303.15, 308.15, and 313.15) K is presented in Table 1, and a change in plot is observed at very low concentrations which is apparent at higher temperatures also in Figure 1. These changes are identified as cmc of surfactant and decreases at higher surfactant concentration. The experimental data of densities, speeds of sound, viscosities, and specific conductivities of solutions of aqueous SDS of molalities between (0.05 and 0.10) mol·kg^{-1} with 1-pentanol and 1-hexanol at T = (298.15 and 308.15) K and atmospheric pressure are reported in Table 2. The uncertainty reported in density measurements is $\pm\ 2\cdot10^{-3}$ kg·m^{-3} and in speed of sound is ± 0.1 m·s^{-1}. In the case of solutions containing 1-pentanol, the experimental values of densities are compared with literature values[9,27] at certain concentrations and different temperatures, and a good agreement is seen. A close inspection of the density data from Table 1 suggests a small change in the slope of density versus alcohol concentration, which gives a slight increase in apparent molar volumes. The sensitivity of densities or apparent molar volumes is far less than for compressibilities, and small, subtle changes may not make a significant contribution to solution densities.[12]

The apparent molar volumes of aqueous SDS solutions as well as that of alcohols (1-pentanol and 1-hexanol) in aqueous

Table 2. Density, ρ, Speed of Sound, u, Viscosity, η, and Specific Conductance, κ, of 1-Pentanol or 1-Hexanol in Aqueous SDS Solutions at (298.15 and 308.15) K

$m/\text{mol}\cdot\text{kg}^{-1}$	$\rho\cdot10^{-3}/\text{kg}\cdot\text{m}^{-3}$	$u/\text{m}\cdot\text{s}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	$\kappa/\text{mS}\cdot\text{cm}^{-1}$	$\rho\cdot10^{-3}/\text{kg}\cdot\text{m}^{-3}$	$u/\text{m}\cdot\text{s}^{-1}$	$\eta/\text{mPa}\cdot\text{s}$	$\kappa/\text{mS}\cdot\text{cm}^{-1}$
		298.15 K				308.15 K		
1-Pentanol				SDS (0.05 $\text{mol}\cdot\text{kg}^{-1}$)[a]				
0.000	0.999078[b]	1498.3	0.986	1.0	0.995942[c]	1520.2	0.790	2.1
0.053	0.998263	1500.0	1.025	1.1	0.995102	1521.2	0.815	2.0
0.062	0.998111	1500.3	1.036	1.0	0.994942	1521.4	0.820	2.0
0.074	0.997938	1500.7	1.045	1.0	0.994750	1521.6	0.825	1.9
0.078	0.997861	1500.8	1.050	0.9	0.994680	1521.7	0.828	1.9
0.104	0.997460[d]	1501.7	1.073	0.8	0.994258[e]	1522.3	0.844	1.9
0.154	0.996675[f]	1503.6	1.114	0.5	0.993427[g]	1523.4	0.876	1.7
0.169	0.996440	1504.2	1.129	0.4	0.993137	1523.8	0.885	1.6
0.184	0.996200	1504.8	1.144	0.3	0.992920	1524.2	0.896	1.5
0.250	0.995145	1506.5	1.200	0.1	0.991790	1524.5	0.940	0.1
1-Pentanol				SDS (0.1 $\text{mol}\cdot\text{kg}^{-1}$)				
0.000	1.000966[h]	1498.0	1.133	2.6	0.997734[i]	1519.1	0.870	3.4
0.070	0.999848	1499.4	1.185	2.6	0.996585	1519.5	0.910	3.3
0.093	0.999490	1499.8	1.204	2.5	0.996208	1519.6	0.926	3.2
0.096	0.999434	1499.9	1.206	2.4	0.996150	1519.6	0.928	3.2
0.106	0.999267[j]	1500.2	1.212	2.4	0.995976[k]	1519.7	0.935	3.1
0.153	0.998507	1501.2	1.243	1.9	0.995169	1520.1	0.962	2.2
0.190	0.997905[l]	1502.1	1.271	1.4	0.994525[m]	1520.5	0.984	1.3
0.250	0.996914	1503.7	1.314	0.2	0.993456	1521.2	1.021	0.2
1-Hexanol				SDS (0.05 $\text{mol}\cdot\text{kg}^{-1}$)				
0.000	0.999078[b]	1498.3	0.986	1.0	0.995942[c]	1520.2	0.790	2.1
0.010	0.998897	1498.4	1.005		0.995758	1520.2	0.797	
0.020	0.998713	1498.4	1.020	1.0	0.995570	1520.1	0.805	1.5
0.030	0.998529	1498.5	1.035	1.0	0.995378	1520.0	0.815	1.5
0.043	0.998287	1498.6	1.054	0.9	0.995125	1519.9	0.828	1.5
0.045	0.998250	1498.6	1.058	0.9	0.995086	1519.9	0.830	1.50
0.060	0.997967	1498.7	1.084	0.5	0.994786	1519.7	0.845	1.0
0.070	0.997780	1498.7	1.099	0.1	0.994586	1519.6	0.855	0.4
0.082	0.997551	1498.8	1.118	0.0	0.994343	1519.5	0.867	0.01
1-Hexanol				SDS (0.1 $\text{mol}\cdot\text{kg}^{-1}$)				
0.000	1.000966[h]	1498.0	1.133	2.6	0.997734[i]	1519.1	0.870	3.4
0.010	1.000779	1498.0	1.140		0.997545	1519.0	0.885	
0.020	1.000590	1498.0	1.147	1.2	0.997351	1518.9	0.893	2.2
0.030	1.000398	1498.0	1.155	1.2	0.997152	1518.8	0.902	2.2
0.040	1.000203	1498.1	1.165	1.2	0.996949	1518.7	0.910	2.2
0.050	1.000008	1498.1	1.176	1.4	0.996746	1518.5	0.918	2.0
0.057	0.999869	1498.1	1.181	1.1	0.996601	1518.5	0.922	1.7
0.062	0.999768	1498.1	1.187	0.9	0.996494	1518.4	0.925	1.3
0.070	0.999611	1498.1	1.193	0.8	0.996328	1518.3	0.931	0.8
0.080	0.999411	1498.1	1.204	0.6	0.996119	1518.2	0.939	0.2

[a] The molal concentration ($\text{mol}\cdot\text{kg}^{-1}$) is given in parentheses for aqueous SDS solutions, and ρ_o and ρ are the densities of aqueous SDS solutions and that containing 1-pentanol, respectively, in $\text{g}\cdot\text{cm}^{-3}$. From ref 9, [b] ($m = 0.0488$), $\rho_o = 0.999014$; [c] ($m = 0.0517$), $\rho_o = 0.996010$; [d] $m = 0.1$, $\rho = 0.997493$; [e] $m = 0.1$, $\rho = 0.994443$; [f] $m = 0.15$, $\rho = 0.996758$; [g] ($m = 0.0517$), $m = 0.15$, $\rho = 0.993656$; [h] ($m = 0.1070$), $\rho_o = 1.001193$; [i] ($m = 0.1123$), $\rho_o = 0.998155$; [j] ($m = 0.1070$), $m = 0.1$, $\rho = 0.995581$; [k] ($m = 0.1123$), $m = 0.1$, $\rho = 0.996474$; [l] $m = 0.2$, $\rho = 0.998007$; [m] ($m = 0.0517$), $m = 0.2$, $\rho = 0.994799$.

SDS solutions can be calculated from the density data using the following expression:

$$\Phi v = (M/\rho) - [\{1000(\rho_o - \rho)\}/m\rho\rho_o] \qquad (1)$$

where ρ_o, M, and m are respectively the density of the pure solvent, the molar mass of the solute, and the molality of the solution in $\text{mol}\cdot\text{kg}^{-1}$. The partial molar volumes, $\Phi^o v$, at infinite dilution of the alcohols in aqueous micelle solution were determined by using the least-squares method to the assumed relation:

$$\Phi v = \Phi^o v + S_v m^{1/2} \qquad (2)$$

The partial molar volume of a solute $\Phi^o v$ reflects the true volume of the solute and the volume change arising from the solute−solvent interactions. It means that the change in $\Phi^o v$ at different surfactant concentrations and temperatures should reflect the changes occurring in its environment in the micellar system. The parameter S_v provides information regarding solute−solute interactions. The derived values of the parameter $\Phi^o v$ and S_v as a function of surfactant concentration and temperature are reported in Table 2.

Thus, the volume of transfer of 1-pentanol or 1-hexanol from water to aqueous surfactant $\Phi^o v(\text{tr})$, was calculated by using the relation:

$$\Phi^o v(\text{tr}) = \Phi^o v\ (\text{in aqueous surfactant}) - \Phi^o v\ (\text{in water}) \qquad (3)$$

where $\Phi^o v$ (in water) is the partial molar volume of the alcohols in water, and its values at 298.15 K have been taken from the literature.[28] The $\Phi^o v(\text{tr})$ values at 298.15 K are summarized in Table 3.

The isentropic compressibility, κ_s, of the solution can be obtained by the Newton−Laplace equation:

Table 3. Fit Coefficients of the Variation of Φv as a Function of the Concentration at (298.15 and 308.15) K for Aqueous SDS−Alcohol Solutions

parameters	298.15 K	308.15 K
$0.05\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Pentanol		
$\Phi^{o}v/\text{cm}^3\cdot\text{mol}^{-1}$	103.34 ± 0.01^{a}	103.87 ± 0.01
$S_v/\text{cm}^3\cdot\text{mol}^{-2}\cdot\text{kg}$	2.09 ± 0.05	3.63 ± 0.03
$\Phi^{o}v_{(\text{water})}/\text{cm}^3\cdot\text{mol}^{-1}$	102.40^{b}	
$\Phi^{o}v_{(\text{tr})}/\text{cm}^3\cdot\text{mol}^{-1}$	0.94	
$0.10\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Pentanol		
$\Phi^{o}v/\text{cm}^3\cdot\text{mol}^{-1}$	103.36 ± 0.01 (104.14^{c})	103.66 ± 0.02
$S_v/\text{cm}^3\cdot\text{mol}^{-2}\cdot\text{kg}$	2.58 ± 0.07	4.64 ± 0.07
$\Phi^{o}v_{(\text{water})}/\text{cm}^3\cdot\text{mol}^{-1}$	102.40^{b}	
$\Phi^{o}v_{(\text{tr})}/\text{cm}^3\cdot\text{mol}^{-1}$	0.96	
$0.05\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Hexanol		
$\Phi^{o}v/\text{cm}^3\cdot\text{mol}^{-1}$	120.09 ± 0.022 (121.14^{c})	120.41 ± 0.03
$S_v/\text{cm}^3\cdot\text{mol}^{-2}\cdot\text{kg}$	3.48 ± 0.11	7.03 ± 0.14
$\Phi^{o}v_{(\text{water})}/\text{cm}^3\cdot\text{mol}^{-1}$	118.65^{d}	
$\Phi^{o}v_{(\text{tr})}/\text{cm}^3\cdot\text{mol}^{-1}$	1.44	
$0.10\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Hexanol		
$\Phi^{o}v/\text{cm}^3\cdot\text{mol}^{-1}$	120.20 ± 0.03 (121.61^{c})	120.56 ± 0.04
$S_v/\text{cm}^3\cdot\text{mol}^{-2}\cdot\text{kg}$	5.14 ± 0.16	8.22 ± 0.17
$\Phi^{o}v_{(\text{water})}/\text{cm}^3\cdot\text{mol}^{-1}$	118.65^{d}	
$\Phi^{o}v_{(\text{tr})}/\text{cm}^3\cdot\text{mol}^{-1}$	1.55	

[a] Standard deviations. [b] Data taken from ref 28. [c] Data taken from ref 12. [d] Data taken from ref 11.

$$\kappa_s = 1/u^2\rho \qquad (4)$$

To further explore the micellar behavior of SDS with monohydric alcohols, viscometric studies were carried out, which are quite sensitive to the shape transitions of colloidal aggregates.[29] The dynamic viscosities, η (reproducibility ± 0.003 mPa·s), of all of the studied solutions at the same concentration as were used for the density and speed of sound measurements are listed in Table 2. The value of the viscosity of 0.3 M solutions of aqueous SDS is 2.0 mPa·s at 298.15 K, whereas at 308.15 K its reported value is 1.2 mPa·s.[30] The relative viscosity (η_{rel}) has been analyzed using the Jones−Dole equation:[31]

$$\eta_{\text{rel}} = \eta/\eta_o = 1 + Am^{1/2} + Bm \qquad (5)$$

where η and η_o are the viscosities of the ternary solutions (alcohol−SDS−water) and binary solvents (SDS−water), respectively, whereas m is the molal concentration of alcohols in ternary solutions. A and B are empirical constants known as viscosity A- and B-coefficients. Coefficient A accounts for the solute−solute interactions and B is a measure of structural modifications induced by the solute−solvent interactions.[32] Equation 5 can be rearranged as:

$$\eta_{\text{rel}} - 1/m^{1/2} = A + Bm^{1/2} \qquad (6)$$

Values of A and B coefficients are obtained from a linear plot of the left-hand side of eq 6 versus $m^{1/2}$ and are listed in Table 4.

Table 4. Viscosity *A*- and *B*-coefficients, Free Energy of Activation of Viscous Flow per Mole of Solvent, $\Delta\mu_1^{0\#}$, and per Mole of Solute, $\Delta\mu_2^{0\#}$, and Limiting Molar Conductance, Λ^0_{m}, of the Alcohols in Aqueous SDS Solutions at T = (298.15 and 308.15) K

parameters	298.15 K	308.15 K
$0.05\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Pentanol		
$A/10^{-1}\ \text{kg}^{1/2}\cdot\text{mol}^{-1/2}$	0.065 ± 0.017	0.016 ± 0.014
$B/10^{-1}\ \text{kg}\cdot\text{mol}^{-1}$	4.701 ± 0.05	5.18 ± 0.04
$\Delta\mu_1^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	9.44	9.20
$\Delta\mu_2^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	647.09	733.60
$\Lambda^0_{\text{m}}/\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$	34.5 ± 3.1	63.8 ± 3.5
$0.10\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Pentanol		
$A/10^{-1}\ \text{kg}^{1/2}\cdot\text{mol}^{-1/2}$	0.048 ± 0.048	-0.038 ± 0.024
$B/10^{-1}\ \text{kg}\cdot\text{mol}^{-1}$	3.973 ± 0.13	4.79 ± 0.07
$\Delta\mu_1^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	9.82	9.48
$\Delta\mu_2^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	542.56	671.15
$\Lambda^0_{\text{m}}/\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$	72.4 ± 5.1	94.5 ± 5.9
$0.05\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Hexanol		
$A/10^{-1}\ \text{kg}^{1/2}\cdot\text{mol}^{-1/2}$	0.661 ± 0.007	0.720 ± 0.006
$B/10^{-1}\ \text{kg}\cdot\text{mol}^{-1}$	3.593 ± 0.035	3.848 ± 0.029
$\Delta\mu_1^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	9.44	9.20
$\Delta\mu_2^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	496.77	547.02
$\Lambda^0_{\text{m}}/\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$	96.0 ± 5.8	115.7 ± 5.4
$0.10\ \text{mol}\cdot\text{kg}^{-1}$ SDS + 1-Hexanol		
$A/10^{-1}\ \text{kg}^{1/2}\cdot\text{mol}^{-1/2}$	0.606 ± 0.003	0.601 ± 0.002
$B/10^{-1}\ \text{kg}\cdot\text{mol}^{-1}$	2.141 ± 0.017	2.681 ± 0.008
$\Delta\mu_1^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	9.82	9.48
$\Delta\mu_2^{0\#}/\text{kJ}\cdot\text{mol}^{-1}$	296.90	379.82
$\Lambda^0_{\text{m}}/\text{S}\cdot\text{cm}^2\cdot\text{mol}^{-1}$	142.3 ± 6.3	207.2 ± 8.4

The viscosity results have also been examined in the light of transition state theory of the relative viscosity proposed by Feakins et al.[33,34] According to this theory, the B-coefficient is given by the relation:

$$B = [\{(\bar{V}_1^0 - \bar{V}_2^0) + \bar{V}_1^0(\Delta\mu_2^{0\#} - \Delta\mu_1^{0\#})/RT\}/1000] \qquad (7)$$

where $\bar{V}_1^0$ is the partial molar volume of the pure solvent (aqueous SDS) and $\bar{V}_2^0$ (= $\Phi^{o}v$) is that for the solute. The free energy of activation per mole of solvent ($\Delta\mu_1^{0\#}$) has been calculated by using the Eyring's viscosity relation:[35]

$$\Delta\mu_1^{0\#} = RT\ln(\eta_0\bar{V}_1^0/hN) \qquad (8)$$

where h and N are Planck's constant and Avogadro number, respectively. Equation 7 rearranges to give the free energy of activation per mole of the solute, $\Delta\mu_2^{0\#}$, as,

$$\Delta\mu_2^{0\#} = \Delta\mu_1^{0\#} + (RT/\bar{V}_1^0)[1000B - (\bar{V}_1^0 - \bar{V}_2^0)] \qquad (9)$$

The values of $\Delta\mu_1^{0\#}$ and $\Delta\mu_2^{0\#}$ are also included in Table 4.

In the case of aqueous SDS solutions the value of apparent molar volumes, Φv, increases slightly and is 237 $\text{cm}^3\cdot\text{mol}^{-1}$ at 298.15 K.[36] In the case of SDBS, the Φv versus $m^{1/2}$ plot shows an increasing trend with an increase in concentration and temperature.[21] A large increase occurs at low concentrations, and a small but well-defined break is observed at higher concentrations.[21,37] In Figure 2 the plots of Φv versus the square root of molality of 1-pentanol or 1-hexanol at different surfactant concentrations and temperatures show that a considerable variation occurs in this property for the two alcohols and depends upon the concentrations of the surfactant and alcohols as well as the temperature. In solutions containing 1-pentanol or 1-hexanol the variations of Φv of studied alcohols are almost linear as shown in Figure 2, parts a and b, respectively.

The partial molar volume of a solute, $\Phi^{o}v$, reflects the volume change arising from the solute−solvent interaction. The change in $\Phi^{o}v$ at different surfactant concentrations and temperatures should reflect the changes occurring in its environment in the micelle system. A perusal of Table 3 reveals that the values of $\Phi^{o}v$ are positive and they increase with the rise in temperature as well as with the increase in concentration of SDS for the studied alcohols in aqueous SDS solutions, suggesting strong solute−solvent interactions. It was shown[11] that $\Phi^{o}v$ dependence of the surfactant concentration is the result of two contributions: the alcohol distribution between the two phases, that is, aqueous and the micellar phase, and the chemical displacement of the micellization equilibrium due to the added alcohol. The latter

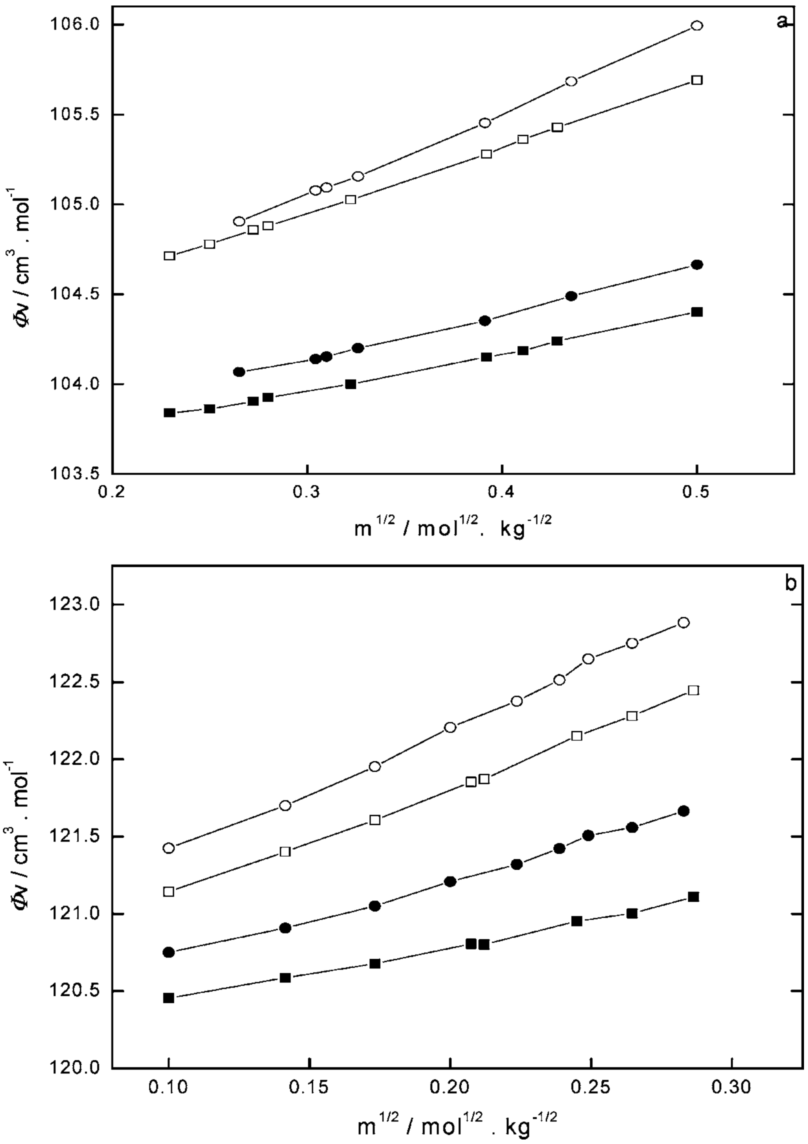

Figure 2. Apparent molar volume, Φv, of aqueous solutions of SDS at different concentrations at 298.15 K: ■, 0.05 mol·kg^{-1}; ●, 0.10 mol·kg^{-1}; and at 308.15 K: □, 0.05 mol·kg^{-1}; ○, 0.10 mol·kg^{-1} with alcohols; (a) 1-pentanol and (b) 1-hexanol.

contribution tends to predominate the longer the alcohol alkyl chain. The value of $\Phi^{o}v$ for 1-pentanol and 1-hexanol in different concentrations of SDS solutions show that there is a significant difference between $\Phi^{o}v$ for both the alcohols and in aqueous data, indicating that the alcohol molecules are partly solubilized in the micelle solution of SDS. The positive sign of S_v indicates that the interaction between the solute species is strong. According to Hoiland et al.,[12] the partial molar volumes of the alcohols in the micellar state are additive with a CH_2 group contribution of 17.1 cm^3·mol^{-1} in nonaqueous solvents. The partial apparent molar properties of transfer provide qualitative as well as quantitative information regarding solute−cosolvent interactions without taking into account the effects of solute−solute interactions.[38,39] A perusal of Table 3 indicates that $\Phi^{o}v$ of alcohols in aqueous SDS are greater than those in pure water, that is, $\Phi^{o}v$ (tr) values are positive for 1-pentanol and 1-hexanol and increase with the increase in surfactant concentration.

The variation of the isentropic compressibility κ_s with the concentration of aqueous SDS has been illustrated in Figure 3. It shows change in slope of κ_s versus $m^{1/2}$ with the change in the speed of sound at different temperatures. The κ_s of aqueous

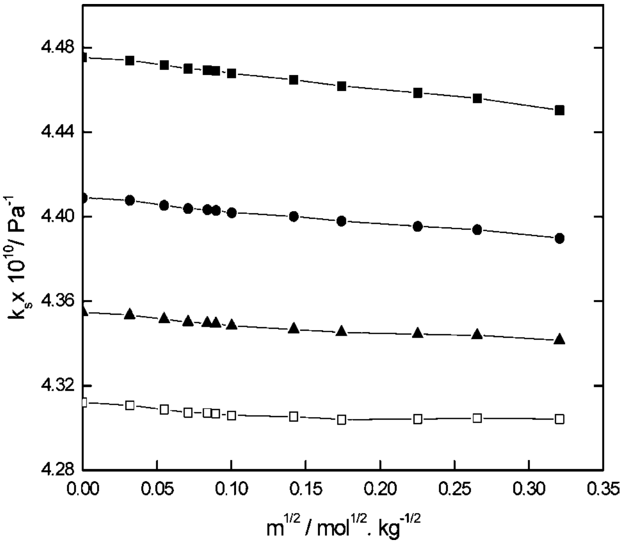

Figure 3. Isentropic compressibility, κ_s, of aqueous solutions of SDS as a function of square root of molality: ■, 298.15 K; ●, 303.15 K; ▲, 308.15 K; and □, 313.15 K.

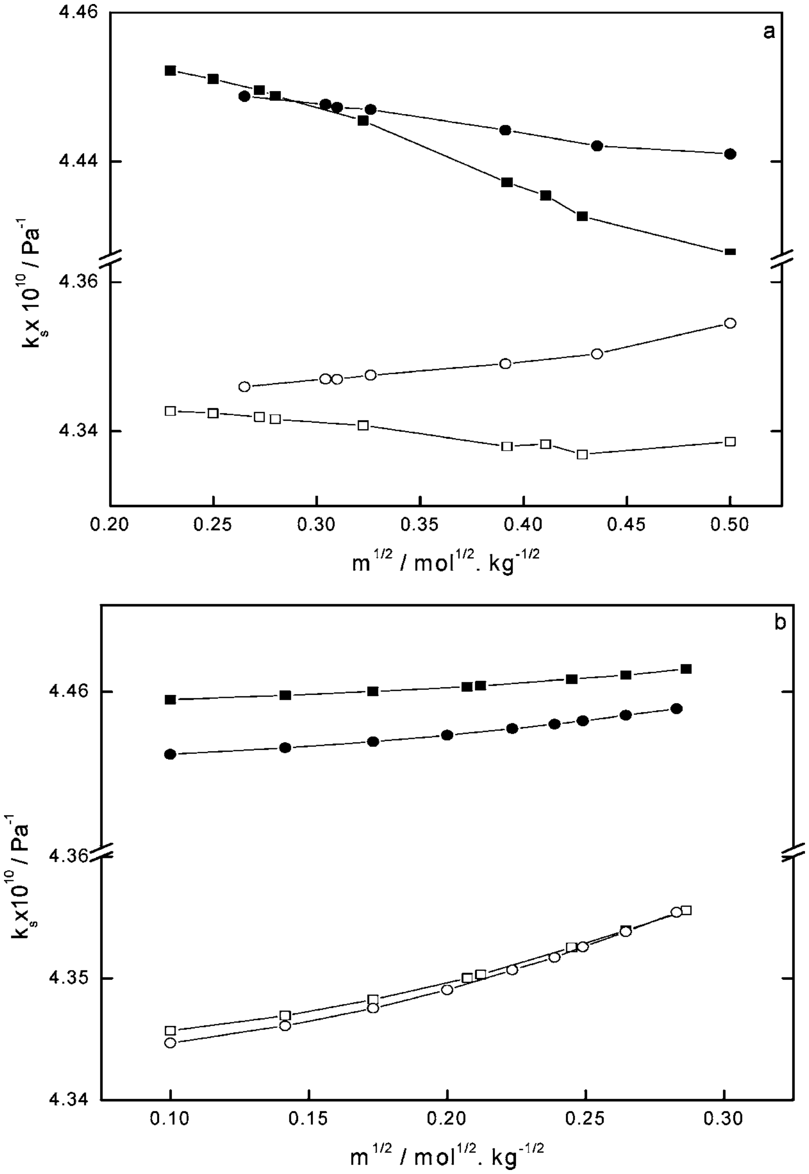

Figure 4. Isentropic compressibility, κ_s, of aqueous solutions of SDS at different concentrations at 298.15 K: ■, 0.05 mol·kg^{-1}; ●, 0.10 mol·kg^{-1}; and at 308.15 K: □, 0.05 mol·kg^{-1}; ○, 0.10 mol·kg^{-1} with alcohols; (a) 1-pentanol and (b) 1-hexanol.

micelle solution of SDS containing 1-pentanol show a decreasing trend except in the case of a solution of 0.10 *m* SDS at 308.15 K as in Figure 4a, whereas in the case of SDS solutions containing 1-hexanol, κ_s values increase at all of the concentrations and temperatures of SDS as given in Figure 4b. The increase in the compressibility may arise because of the decrease in the structured water as a result of the transfer of the additive alcohols from the aqueous phase to the micelle aggregates.[37] This change is compensated by the loss of free space in the micelle interior upon addition of alcohols. It results in an increase in the compressibility of the micellar solutions. The decrease in κ_s, as in case of 1-pentanol, suggests that it is incompletely transferred from the aqueous environment to the micelle or it is located near the surface of the micelle. According to Gonzalez-Perez et al.,[40] hydration makes a negative contribution to the compressibility of a solute, as observed for monomeric surfactants as well as for simple electrolytes or their ions. According to Hoiland et al.,[12] a plot of density or compressibility versus alcohol concentration in aqueous SDS–hexanol solutions was found to be significantly curved at low alcohol concentrations, typically below 0.04 mol·kg^{-1}. The pronounced curvature at low alcohol concentrations is mainly an effect of the changes in cmc. However, above this molal concentration a region of almost linear relationship is observed as in present study.

The values of viscosity *A*- and *B*-coefficients, obtained from a linear plot of the left-hand side of eq 6 versus $m^{1/2}$, are listed in Table 3. The relative viscosities, at different concentrations of aqueous SDS as a function of molal concentration of 1-pentanol or 1-hexanol, are linear and show an increasing trend with the increase in the concentration of alcohols. Table 4 shows that the values of *B*-coefficients are large and positive in aqueous SDS solutions containing 1-pentanol or 1-hexanol, suggesting strong SDS–water–alcohol interactions. However, the values of *A*-coefficients are small and positive in the studied systems except in case of 0.10 *m* SDS–1-pentanol solution, where it was found to be negative, indicating weak alcohol–alcohol interactions. The increasing value of *B*-coefficients and a reverse trend in *A*-coefficients with the rise in temperature are seen. Because of the complex nature of *A*-coefficients, it is not discussed in detail. The *B*-coefficients measure the size and shape effects as well as the structural effect induced by solute–solvent interaction.[29] The positive values of *B*-coef-

Table 5. Chemical Shift (*δ*) of Protons Close to the Tail of SDS in the Presence and in the Absence of 1-Pentanol in SDS (0.05 and 0.10) mol·kg^{-1} at $T = 295$ K

SDS	1-pentanol	proton peaks in SDS micellar solutions/ppm	
mol·kg^{-1}	mol·kg^{-1}	peak c	peak d
0.05	0.0	1.6127	3.9485
	0.05	1.5749	3.9173
	0.106	1.5740	3.9159
	0.208	1.5693	3.9053
0.10	0.0	1.6129	3.9491
	0.068	1.5828	3.9224
	0.105	1.5811	3.9196
	0.245	1.5753	3.9132

ficients are associated with a structure-making phenomenon. The thermodynamic investigations for aqueous SDS micellar solutions at different concentrations containing alcohols clearly indicate that the studied alcohol molecules behave as structure makers.

The computed values of the free energy of activation per mole of solvent ($\Delta\mu_1^{0\#}$) and that per mole of solute ($\Delta\mu_2^{0\#}$) is listed in Table 4. It is evident from Table 4 that for all of the studied systems, the $\Delta\mu_2^{0\#}$ values are positive and much larger than those of $\Delta\mu_1^{0\#}$ values. This indicates that the interactions between alcohols and solvent (aqueous SDS) molecules in the ground state are stronger than in the transition state. Hence in the transition state, the solvation of the solute molecules is less favored in free energy terms. The $\Delta\mu_2^{0\#}$ values increase with an increase in temperature for the studied alcohols, indicating that the solute–solvent interactions increase with rise in temperature. Thus, the conclusions drawn from $\Delta\mu_2^{0\#}$ are in agreement with those drawn from the trends of other thermodynamic parameters studied. All of the thermodynamic studies clearly indicate the strong solute–solvent interactions. The volumetric and viscometric studies are in good agreement with each other.

The limiting molar conductance, Λ^0_m, has been regarded as a measure of alcohol–SDS–water interactions.[41,42] The Λ^0_m values for alcohols in aqueous SDS solutions were obtained by extrapolating the linear plots of molar conductance, Λ_m versus $m^{1/2}$, to zero concentration. The Λ^0_m values for alcohols in aqueous SDS solutions are incorporated in Table 4. The large and positive values of Λ^0_m of alcohols in aqueous SDS solutions shows an increasing trend with the increase in temperature. It clearly indicates that alcohol–surfactant–water interactions are stronger and increase with an increase in temperature. The values of Λ^0_m for 1-hexanol are greater than those for 1-pentanol indicating relatively strong 1-hexanol–SDS–water interactions.

The ^{1}H NMR chemical shift studies of solutions containing (0.05 and 0.10) mol·kg^{-1} SDS and different concentrations of 1-pentanol are reported in Table 5. Typical spectra for 0.05 mol·kg^{-1} aqueous SDS and the assignment of various peaks in the absence and in the presence of 1-pentanol (0.05 mol·kg^{-1}) are shown in Figure 5. A careful inspection of various ^{1}H signals in the case of pure surfactant solution and a shift in the position of these signals upon mixing 1-pentanol helps to deduce the preferential solubilization sites of 1-pentanol in aqueous SDS solution. A comparison of the chemical shift (*δ*) of the proton spectra of aqueous SDS solutions (a), with that containing 1-pentanol (b), clearly indicates that the site for preferential solubilization of alcohol is close to the headgroup of the micelle. This is apparent from the significant change in frequencies of the proton peaks (peaks c and d) of Table 4. In SDS solutions at the studied concentrations (0.05 and 0.10 mol·kg^{-1}) peaks c and d show a small but upfield shift with an increase in the concentration of 1-pentanol.

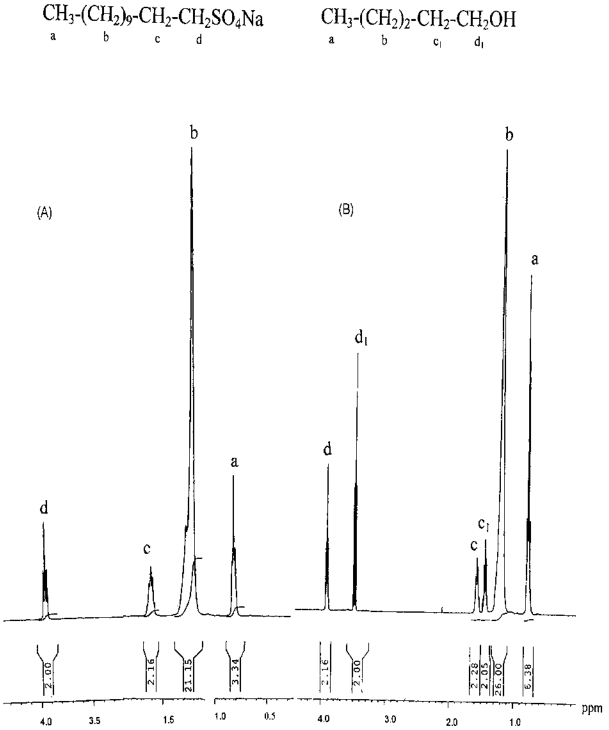

Figure 5. ^{1}H NMR spectra in ppm at 295 K: (a) 0.05 *m* SDS; (b) 0.05 *m* 1-pentanol in 0.05 *m* SDS in D_2O.

The peaks for the proton close to the tail of SDS are overlapping with the peaks due to the methylene protons of 1-pentanol, which makes it difficult to interpret the shift in the position of protons close to the tail of SDS of NMR spectra. Apart from this, the presence of a large number of protons (close to the tail of SDS) appearing over a narrow range of chemical shift makes their exact assignment unrealistic. Since the change of chemical shift caused by the aggregation of molecules is usually related to structural changes,[5] the NMR experiments imply that the location of the added alcohols is in the interfacial region of micellar aggregates. The results of the present study agree well with those reported by Reekmans et al.,[5] where they studied ^{13}C NMR spectra of aqueous SDS–butanol/heptanol systems. Similar observations were found in case of aqueous SDBS–1-pentanol solutions, where the preferable site for the solubilization of 1-pentanol was the aromatic ring of the micelle.[21]

Conclusion

The thermodynamic investigations for aqueous SDS micellar solutions at different concentrations containing alcohols clearly indicate the strong alcohol–surfactant interaction concluded from the studied partial molar property. The studied alcohol molecules behave as structure makers as concluded from viscosity *B*-coefficient values. The site for preferential solubilization of the alcohol is close to the headgroup of the micelle, as interpreted from NMR studies. The conclusion seems to be that alcohol solubilization at low surfactant concentration is a two-step mechanism. The first is generally accepted that the alcohol molecules presumably reside in the palisade layer of the micelle, and the second is that the alcohol molecules remain parallel to the highly oriented surfactant molecules. At higher concentrations of alcohols, the solubilization site becomes more hydrophobic. Presumably, it means that some of the alcohol molecules become solubilized in the micellar interior, experiencing a hydrophobic environment, while some still remain in the palisade layer. A study of such systems at higher surfactant concentrations will help to deduce the approximate fraction of alcohol molecules that is solubilized in the micellar interior for aqueous SDS/medium chain alcohols.

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