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Densities and partial molar volumes of dodecyltrimethylammonium bromide in binary systems (methanol+water) at T=(298.15 to 323.15) K

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Abstract: The densities of dodecyltrimethylammonium bromide in pure water and in methanol + water mixed solvent media containing (0.10, 0.20, 0.30, and 0.40) volume fractions of methanol were measured at (298.15, 308.15, 318.15, and 323.15) K. The concentrations are varied from (0.4×10^{-1} to 1.2×10^{-1}) mol kg^{-1}. The results showed almost increase on the densities with increasing surfactant concentration. Also, the densities are found to decrease with increasing temperature over the entire concentration range investigated in a given mixed solvent medium and these values are found to decrease with increasing methanol content in the solvent composition. The effects of concentration, solvent composition and temperature on the partial molar volumes are discussed.

Keywords: DTAB, Solvent Composition, Methanol-Water Mixed Solvent Media

1. Introduction

Various thermodynamic parameters have been studied in aqueous organic mixed solvent media and the effect of organic solvents have been discussed in literatures (1, 2, 3, 4). One of the research articles in aqueous organic mixed solvent media from our group on partial molar volumes of anionic surfactant sodium lauryl sulphate has been published (5). The experimental procedures have been used to calculate the partial molar volume for ionic and non-ionic organic compounds in aqueous solutions (6, 7). However, the evaluation of these procedures for surfactant is limited by the lack of availability of reliable experimental data of a broad variety of chemical structures and macromolecular characteristics. We have found the explanation of the partial molar volume in a number of papers in details (8, 9, 10). Hence, only basic relation will be used on our system to calculate the partial molar volume. The partial molar volume, V_B , is defined by the following equation;

$$V_B = (\partial V / \partial n)_{T,p} \quad (1)$$

Where, ∂V represent change in total volume and n as the number of moles. The partial molar volume is often provided in units of partial molar volume (cm^3 / mol). If there is concentration dependence, the partial molar volumes have to be extrapolated to concentration zero using one of the following two equations which calculate the apparent volume at the finite concentrations, C (10, 11)

$$v = \frac{1}{\rho_0} - \frac{1}{c}(\frac{\rho}{\rho_0} - 1) \quad (2)$$

With C in g cm^{-3} or

$$V_B = \frac{M}{\rho_0} - \frac{10^3}{c}(\frac{\rho}{\rho_0} - 1) \quad (3)$$

where, M is the molecular weight of the dodecyltrimethylammonium bromide, ρ_0 is the density of the solvent, ρ is the density of the solution and C is equivalent concentration in mol kg^{-1}.

In order to calculate partial molar volumes, the solution densities are thoroughly measured for dodecyltrimethylammonium bromide at the temperatures (298.15, 308.15, 318.15, and 323.15) K in pure water and in methanol + water mixed solvent media containing (0.10, 0.20, 0.30, and 0.40) volume fractions of methanol.

2. Experimental Investigations

2.1. Chemicals and Materials

Dodecyltrimethylammonium bromide was purchased from Loba Chemie Private Limited, Mumbai, India whereas methanol was purchased from Merck, India and was distilled with phosphorous pentoxide and then redistilled over calcium hydride. The purified solvent had a density of 0.77723 g cm^{-3} and a co-efficient of viscosity of 0.47424 mPa s at 308.15 K; these values are in good agreement with the literature values (12). Triply distilled water with a specific conductance less than 10^{-6} S cm^{-1} at 308.15 K was used for the preparation of the mixed solvents. The physical properties of methanol + water mixed solvents used in this study are shown in Table 1 and those values are matched with the published works (5, 13, 14).

Table 1. *Properties of methanol +water mixtures containing (0.10, 0.20, 0.30, and 0.40) volume fraction of methanol at T= (298.15, 308.15, 318.15, and 323.15) K*

T/K	ρ_0 **/(g cm^{-3})**	η_0 **/ (mPa s)**	***D***
$\varphi_1 = 0.10$			
298.15	0.98297	1.0844	75.09
308.15	0.97973	0.8665	71.57
318.15	0.97604	0.7017	68.18
323.15	0.97438	0.6375	66.45
$\varphi_1 = 0.20$			
298.15	0.96963	1.3106	71.61
308.15	0.96632	1.0217	68.14
318.15	0.96162	0.8075	64.80
323.15	0.95875	0.7300	63.15
$\varphi_1 = 0.30$			
298.15	0.95620	1.4712	67.65
308.15	0.95160	1.1418	64.25
318.15	0.94626	0.8957	60.99
323.15	0.94331	0.8052	59.41
$\varphi_1 = 0.40$			
298.15	0.93957	1.4475	63.53
308.15	0.93364	1.2034	60.34
318.15	0.93140	0.9309	57.18
323.15	0.92800	0.8288	55.62

2.2. Density Measurements

To calculate the partial molar volume of dodecyltrimethyl-ammonium bromide in pure water, the density of pure water was used from the literatures (15, 16). The relative permittivity of methanol + water mixtures at the experimental temperatures were obtained by regressing the relative permittivity data as function of solvent composition from the literature (17).

The pycnometric method was used for measuring the density. The stock solutions were freshly prepared for each concentration series to avoid problems of aging and microorganism contamination, which was found to occur with diluted surfactant solutions (18).

The densities of solutions were determined by the use of Ostwald-Sprengel type pycnometer of about 25 cm^3 capacity. The sample solution was transfused into the pycnometer by using a medical syringe. The pycnometer was then tightly fixed in a thermostat at the experimental temperatures within ± 0.005 K. After thermal equilibrium was attained, the mass of the pycnometer was measured with electronic balance and the density was calculated. Density measurements are precise within ±0.00005 g cm^{-3}, which is satisfactory for our purpose. In order to avoid moisture pickup, all solutions were prepared in a dehumidified room with utmost care. In all cases, the experiments were performed in three replicates. The partial molar volumes at different molalities of the solutions are given in table 2 along with the standard error at 95 % confidence interval.

3. Results and Discussions

Mixing methanol and water is exothermic as well as they occupy less volume than the sum of their volumes before mixing. The mixture was thoroughly shaken, and kept 24 hours for the released air bubbles to escape before attempting to make the solution of dodecyltrimethyl-ammonium bromide. Methanol + water is a popular mixed solvent and has been extensively studied (19). When methanol and water are mixed, the density is decreased with the increase of methanol content for the methanol + water mixed solvent system (Table 1).

The densities for the dodecyltrimethylammonium bromide in pure water and in four different methanol-water mixtures (containing 0.1, 0.2, 0.3, and 0.4 volume fractions of methanol) at 298.15, 308.15, 318.15, and 323.15 K are depicted in Figures 1- 4 which show the variation of densities of the investigated solutions as a function of the surfactants concentration. From these Figures, it is evident that the densities exhibits almost increase with increasing concentration within the concentration range investigated here.

However, the density of the system increases with the addition of surfactant (Table 2). This behaviour has been found to be similar in the literatures (5, 20, 21, 22). It was also seen for density values for surfactants decrease with increasing temperature on Chauhan et al. work (22). Also, our density data for pure water of dodecyltrimethylammonium bromide match with R. De Lisi et al. Work (16).

Obviously, the concentration dependence of density follows the same pattern at all the temperatures and solvent composition investigated. In fact, the variations of density with DTAB concentrations are always found to be linear. We, therefore, determined the density of the solvent, by extrapolating the density values to zero DTAB concentration. It is very interesting to see our results of density of the solvent from Table 2 and calculated from the graph, Table 3 which is almost matching with each other. This shows that the density data of dodecyltrimethylammonium bromide in pure water and methanol -water mixed solvent media looks correct.

The slopes of density versus DTAB concentration graph are always found to be positive in methanol–water mixtures, indicating strong ion-ion interactions in these media. Moreover, the slopes are found to increase in the mixed solvent media become richer in methanol, irrespective of temperature indicating greater counterion binding with increasing amount of methanol in the solvent mixtures.

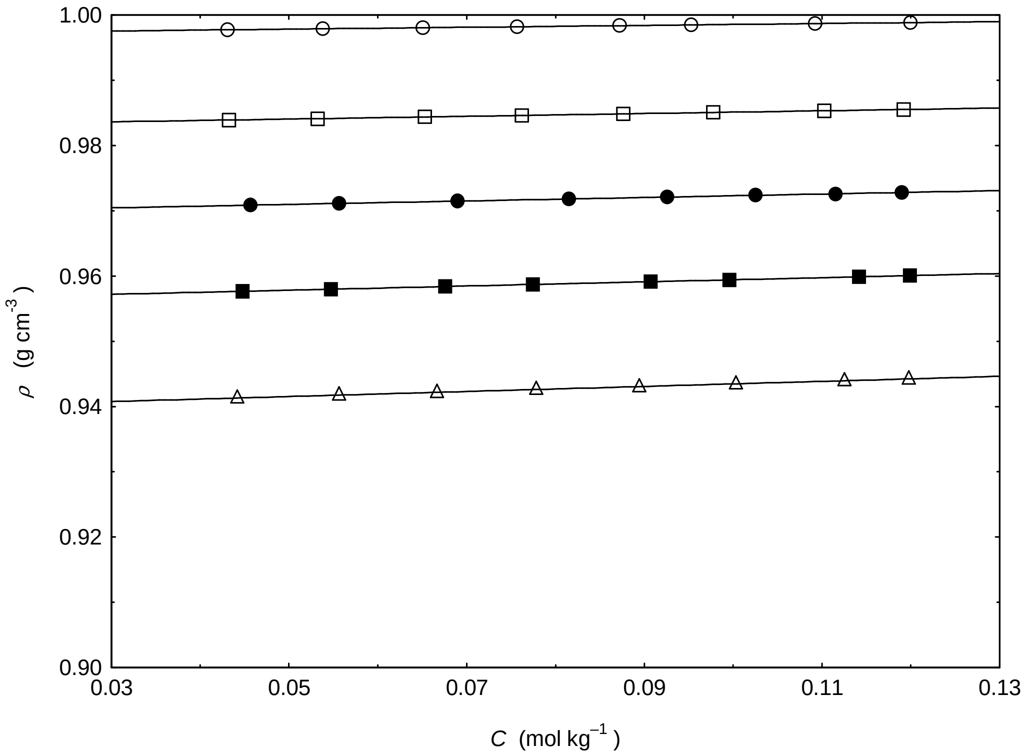

Figure 1. *Concentration dependence of density for dodecyltrimethylammonium bromide at 298.15 K, in pure water (open circles) and different methanol (1) + water (2) mixtures (open squares, 0.10 methanol; closed circles, 0.20 methanol; closed squares, 0.30 methanol; triangles, 0.40 methanol).*

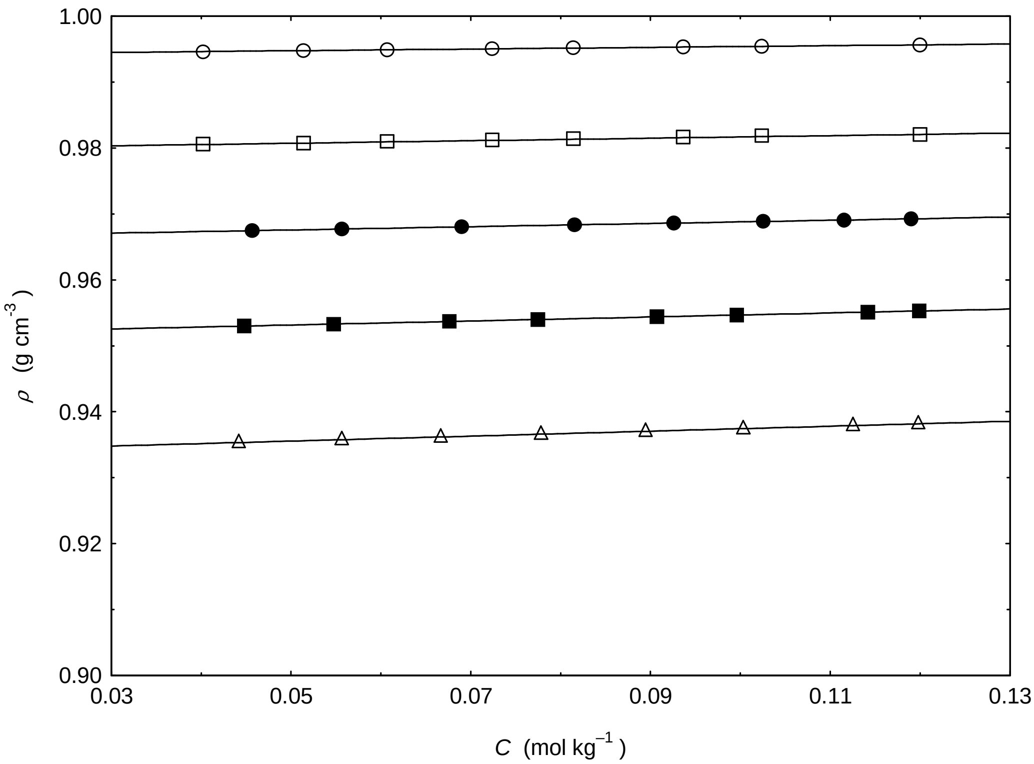

Figure 2. *Concentration dependence of density for dodecyltrimethylammonium bromide at 308.15 K, in pure water (open circles) and different methanol (1) + water (2) mixtures (open squares, 0.10 methanol; closed circles, 0.20 methanol; closed squares, 0.30 methanol; triangles, 0.40 methanol).*

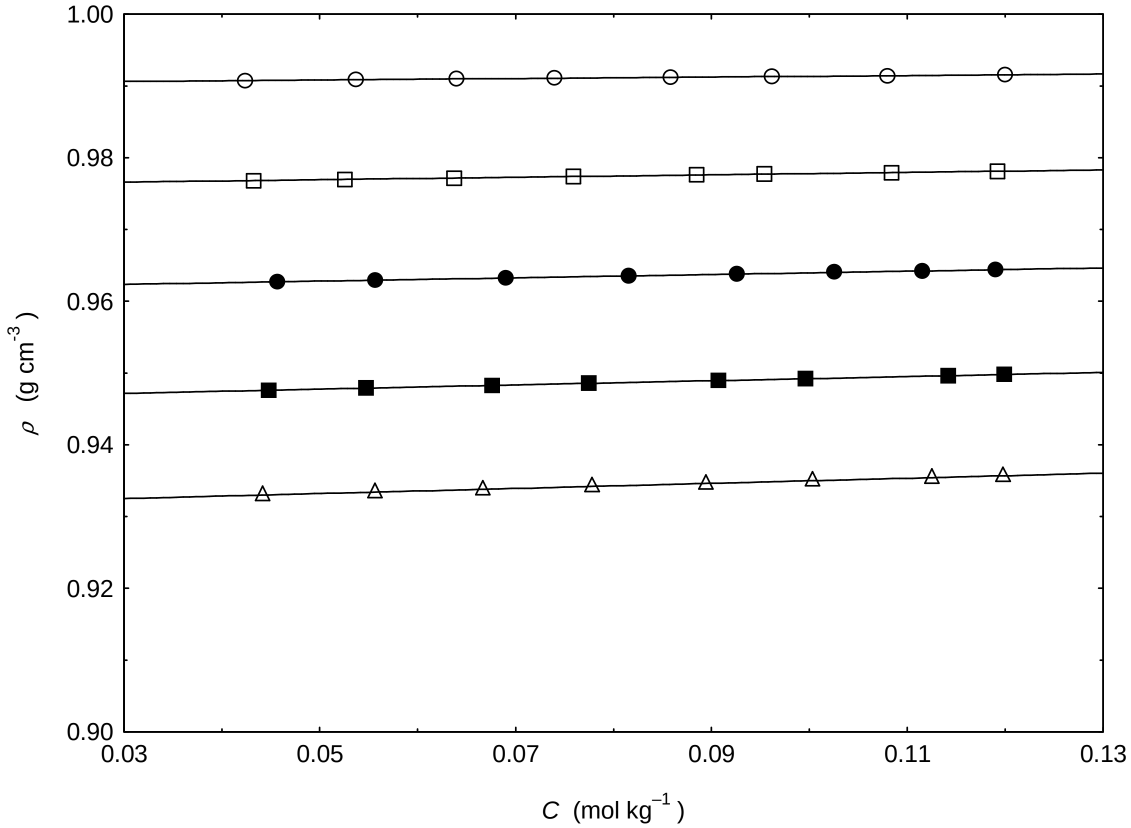

Figure 3. *Concentration dependence of density for dodecyltrimethylammonium bromide at 318.15 K, in pure water (open circles) and different methanol (1) + water (2) mixtures (open squares, 0.10 methanol; closed circles, 0.20 methanol; closed squares, 0.30 methanol; triangles, 0.40 methanol).*

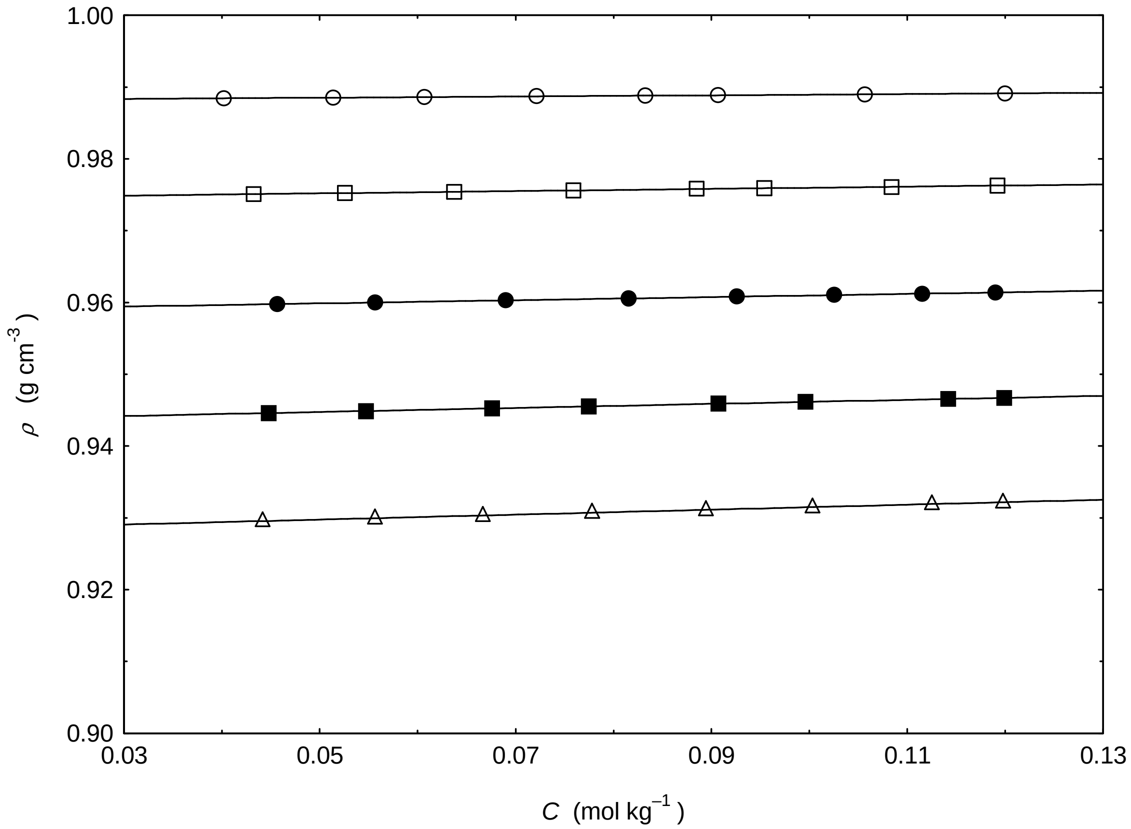

Figure 4. *Concentration dependence of density for dodecyltrimethylammonium bromide at 298.15 K, in pure water (open circles) and different methanol (1) + water (2) mixtures (open squares, 0.10 methanol; closed circles, 0.20 methanol; closed squares, 0.30 methanol; triangles, 0.40 methanol).*

Table 2. *Concentration, density and partial molar volume of dodecyltrimethylammonium bromide in pure water and methanol + water mixed solvent media at T = (298.15, 308.15, 318.15, and 323.15) K*

C /(mol kg^{-1})	ρ /(g cm^{-3})	V_B /(cm^{3}mol^{-1})
T = 298.15 K		
ϕ_1 = 0.00		
0.12000	0.99884	294.45 ± 0.21
0.10926	0.99870	294.32 ± 0.12
0.09530	0.99851	294.09 ± 0.26
0.08725	0.99840	293.94 ± 0.29
0.07569	0.99821	294.12 ± 0.34
0.06508	0.99806	293.97 ± 0.40
0.05384	0.99792	293.41 ± 0.46
0.04311	0.99775	293.41 ± 0.58
ϕ_1 = 0.10		
0.11921	0.98552	291.91 ± 0.22
0.11028	0.98535	291.73 ± 0.23
0.09775	0.98509	291.62 ± 0.25
0.08763	0.98486	291.74 ± 0.29
0.07620	0.98462	291.67 ± 0.34
0.06530	0.98441	291.26 ± 0.35
0.05324	0.98412	291.71 ± 0.40
0.04326	0.98392	291.37 ± 0.60
ϕ_1 = 0.20		
0.11900	0.97280	290.54 ± 0.22
0.11154	0.97258	290.71 ± 0.24
0.10256	0.97242	289.93 ± 0.25
0.09260	0.97212	290.27 ± 0.28
0.08156	0.97181	290.45 ± 0.32
0.06898	0.97150	290.02 ± 0.37
0.05566	0.97113	290.22 ± 0.46
0.04569	0.97086	290.24 ± 0.56
ϕ_1 = 0.30		
0.11990	0.96006	288.78 ± 0.22
0.11417	0.95988	288.76 ± 0.23
0.09960	0.95942	288.66 ± 0.26
0.09072	0.95916	288.34 ± 0.29
0.07746	0.95872	288.44 ± 0.34
0.06760	0.95840	288.42 ± 0.40
0.05473	0.95798	288.45 ± 0.47
0.04478	0.95766	288.38 ± 0.58
ϕ_1 = 0.40		
0.11980	0.94423	286.78 ± 0.22
0.11253	0.94395	286.75 ± 0.24
0.10032	0.94349	286.57 ± 0.27
0.08945	0.94306	286.66 ± 0.30
0.07782	0.94263	286.32 ± 0.34
0.06666	0.94217	286.65 ± 0.40
0.05562	0.94176	286.28 ± 0.48
0.04416	0.94130	286.46 ± 0.60
T = 308.15 K		
ϕ_1 = 0.00		
0.12000	0.99563	297.19 ± 0.21
0.10238	0.99540	297.21 ± 0.25
0.09363	0.99533	296.73 ± 0.27
0.08140	0.99521	296.21 ± 0.31
0.07237	0.99506	296.57 ± 0.35
0.06068	0.99491	296.41 ± 0.42
0.05138	0.99478	296.45 ± 0.50
0.04021	0.99460	297.13 ± 0.65
ϕ_1 = 0.10		
0.11921	0.98204	294.97 ± 0.29
0.11028	0.98189	294.73 ± 0.23
0.09775	0.98166	294.58 ± 0.27
0.08763	0.98144	294.79 ± 0.30
0.07620	0.98123	294.65 ± 0.35
0.06530	0.98104	294.26 ± 0.39
0.05324	0.98077	294.76 ± 0.48
0.04326	0.98059	294.43 ± 0.58
ϕ_1 = 0.20		
0.11900	0.96925	293.60 ± 0.22
0.11154	0.96906	293.67 ± 0.23
0.10256	0.96891	292.94 ± 0.26
0.09260	0.96863	293.28 ± 0.28
0.08156	0.96834	293.46 ± 0.32
0.06898	0.96806	292.98 ± 0.37
0.05566	0.96771	293.24 ± 0.46
0.04569	0.96746	293.27 ± 0.56
ϕ_1 = 0.30		
0.11990	0.95528	291.77 ± 0.22
0.11417	0.95510	291.80 ± 0.23
0.09960	0.95467	291.65 ± 0.27
0.09072	0.95442	291.36 ± 0.35
0.07746	0.95400	291.46 ± 0.34
0.06760	0.95370	291.39 ± 0.39
0.05473	0.95330	291.41 ± 0.50
0.04478	0.95299	291.33 ± 0.90
ϕ_1 = 0.40		
0.11980	0.93817	289.77 ± 0.23
0.11253	0.93789	289.79 ± 0.24
0.10032	0.93745	289.57 ± 0.27
0.08945	0.93703	289.67 ± 0.30
0.07782	0.93662	289.26 ± 0.36
0.06666	0.93617	289.62 ± 0.40
0.05562	0.93577	289.27 ± 0.49
0.04416	0.93532	289.48 ± 0.61
T = 318.15 K		
ϕ_1 = 0.00		
0.12000	0.99154	300.45 ± 0.22
0.10799	0.99140	300.53 ± 0.23
0.09619	0.99132	300.04 ± 0.27
0.08582	0.99120	299.99 ± 0.30
0.07396	0.99110	299.62 ± 0.35
0.06396	0.99098	299.70 ± 0.40
0.05369	0.99087	299.52 ± 0.47
0.04237	0.99073	299.67 ± 0.62
ϕ_1 = 0.10		
0.11921	0.97810	298.20 ± 0.22
0.10839	0.97791	298.24 ± 0.24
0.09539	0.97773	297.78 ± 0.27
0.08850	0.97762	297.62 ± 0.29
0.07590	0.97738	297.82 ± 0.34
0.06373	0.97717	297.75 ± 0.40
0.05256	0.97697	297.76 ± 0.49
0.04326	0.97681	297.71 ± 0.61
ϕ_1 = 0.20		
0.11900	0.96437	296.60 ± 0.22
0.11154	0.96419	296.69 ± 0.23
0.10256	0.96406	295.91 ± 0.25
0.09260	0.96379	296.28 ± 0.28

C /(mol kg^{-1})	ρ /(g cm^{-3})	V_B /(cm^{3}mol^{-1})
0.08156	0.96352	296.44 ± 0.33
0.06898	0.96326	295.94 ± 0.38
0.05566	0.96293	296.19 ± 0.47
0.04569	0.96269	296.28 ± 0.57
ϕ_1 = 0.30		
0.11990	0.94979	294.75 ± 0.23
0.11417	0.94962	294.76 ± 0.23
0.09960	0.94920	294.67 ± 0.27
0.09072	0.94896	294.39 ± 0.30
0.07746	0.94856	294.46 ± 0.34
0.06760	0.94827	294.42 ± 0.39
0.05473	0.94789	294.41 ± 0.50
0.04478	0.94759	294.45 ± 0.59
ϕ_1 = 0.40		
0.11980	0.93567	292.79 ± 0.22
0.11253	0.93542	292.70 ± 0.24
0.10032	0.93500	292.53 ± 0.27
0.08945	0.93460	292.69 ± 0.16
0.07782	0.93421	292.29 ± 0.35
0.06666	0.93379	292.58 ± 0.42
0.05562	0.93341	292.27 ± 0.49
0.04416	0.93299	292.41 ± 0.61
T = 323.15 K		
ϕ_1 = 0.00		
0.12000	0.98910	302.97 ± 0.21
0.10567	0.98898	302.88 ± 0.24
0.09069	0.98890	302.25 ± 0.28
0.08325	0.98883	302.22 ± 0.31
0.07216	0.98873	302.12 ± 0.35
0.06068	0.98862	302.09 ± 0.42
0.05138	0.98853	302.03 ± 0.49
0.04021	0.98841	302.25 ± 0.63
ϕ_1 = 0.10		
0.11921	0.97627	300.18 ± 0.21
0.10839	0.97609	300.26 ± 0.24
0.09539	0.97593	299.79 ± 0.27
0.08850	0.97583	299.64 ± 0.29
0.07590	0.97561	299.82 ± 0.34
0.06373	0.97542	299.73 ± 0.42
0.05256	0.97524	299.68 ± 0.49
0.04326	0.97509	299.64 ± 0.61
ϕ_1 = 0.20		
0.11900	0.96138	298.57 ± 0.23
0.11154	0.96120	298.69 ± 0.23
0.10256	0.96108	297.91 ± 0.25
0.09260	0.96082	298.29 ± 0.28
0.08156	0.96056	298.47 ± 0.32
0.06898	0.96031	298.01 ± 0.37
0.05566	0.95999	298.33 ± 0.49
0.04569	0.95977	298.23 ± 0.85
ϕ_1 = 0.30		
0.11990	0.94671	296.80 ± 0.22
0.11417	0.94655	296.78 ± 0.23
0.09960	0.94615	296.66 ± 0.27
0.09072	0.94592	296.37 ± 0.29
0.07746	0.94553	296.47 ± 0.35
0.06760	0.94525	296.44 ± 0.39
0.05473	0.94488	296.47 ± 0.48
0.04478	0.94460	296.37 ± 0.62
ϕ_1 = 0.40		
0.11980	0.93217	294.77 ± 0.23
0.11253	0.93192	294.72 ± 0.24
0.10032	0.93151	294.56 ± 0.27
0.08945	0.93112	294.68 ± 0.30
0.07782	0.93074	294.32 ± 0.35
0.06666	0.93033	294.61 ± 0.40
0.05562	0.92996	294.30 ± 0.48
0.04416	0.92955	294.44 ± 0.61

Table 3. *Density of the solvent (ρ_0), experimental slopes and the correlation coefficients of fits (as r^2) of dodecyltrimethylammonium bromide from Figures 1 to 4 in pure water and methanol-water mixtures at 298.15, 308.15, 318.15 and 323.15 K.*

Vol. of methanol	slope	ρ_0	r^2
T = 298.15 K			
0	0.014	0.99714	0.9992
0.1	0.021	0.98301	0.9996
0.2	0.026	0.96966	0.9989
0.3	0.032	0.95624	0.9998
0.4	0.039	0.93960	0.9999
T = 308.15 K			
0	0.013	0.99413	0.9933
0.1	0.019	0.97976	0.9993
0.2	0.024	0.96635	0.9987
0.3	0.030	0.95164	0.9998
0.4	0.037	0.93367	0.9998
T = 318.15 K			
0	0.010	0.99032	0.9961
0.1	0.017	0.97608	0.9986
0.2	0.023	0.96166	0.9998
0.3	0.029	0.94629	0.9998
0.4	0.036	0.93143	0.9999
T = 323.15			
0	0.009	0.98809	0.9911
0.1	0.015	0.97443	0.9985
0.2	0.022	0.95877	0.9983
0.3	0.028	0.94335	0.9998
0.4	0.035	0.92803	0.9999

The effects of temperature and relative permittivity on the densities values are directly evident from Figures 1 - 4. At each temperature, the densities values are found to decrease with decreasing relative permittivity in going from 0.1 volume fractions of methanol to 0.4 volume fractions of methanol over the entire concentration range investigated. An increase in temperature, on the other hand, is found to decrease the density in a given solvent medium as manifested in these Figures. Evaluation of the solvent density lead to important insight as to the solution behavior of dodecyltrimethylammonium bromide. The solvent density values thus obtained along with the slopes and the correlation coefficients of fits, (as r^2) are listed in Table 3.

Furthermore, at a given temperature, slopes are found to increase whereas the solvent density values are found to decrease as the solvent medium gets richer in methanol. Also, in all compositions; the solvent density, on the other hand, found to decrease with temperature (Table 3).

The partial molar volumes for the dodecyltrimethylammonium bromide in pure water and four other different methanol + water mixtures containing (0.10, 0.20, 0.30, and 0.40) volume fraction of methanol at (298.15, 308.15, 318.15, and 323.15) K are shown in Table 2. Figures 5 to 8 show the variation of partial molar volumes of the investigated solution as a function of the surfactant concentration. From these Figures, the partial molar volumes exhibit almost independent with increasing concentration within the examined concentration ranges in this study.

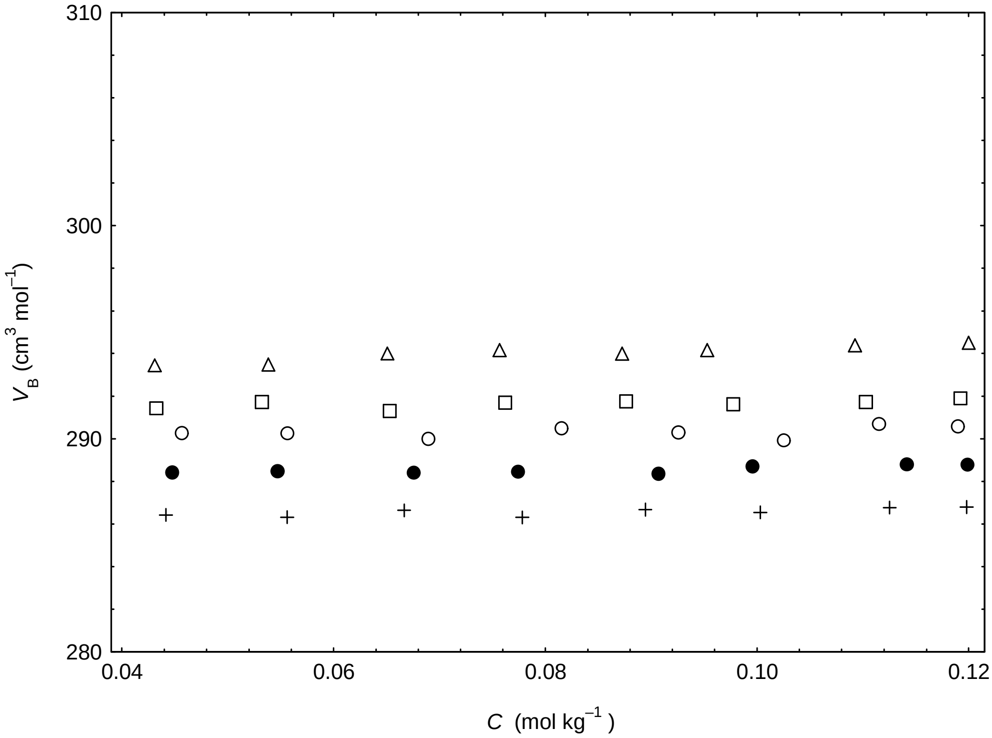

Figure 5. *Concentration independence of partial molar volume for dodecyltrimethylammonium bromide at 298.15 K, in pure water (triangles) and different methanol (1) + water (2) mixtures (squares, 0.10 methanol; circles, 0.20 methanol; closed circles, 0.30 methanol; crosses, 0.40 methanol).*

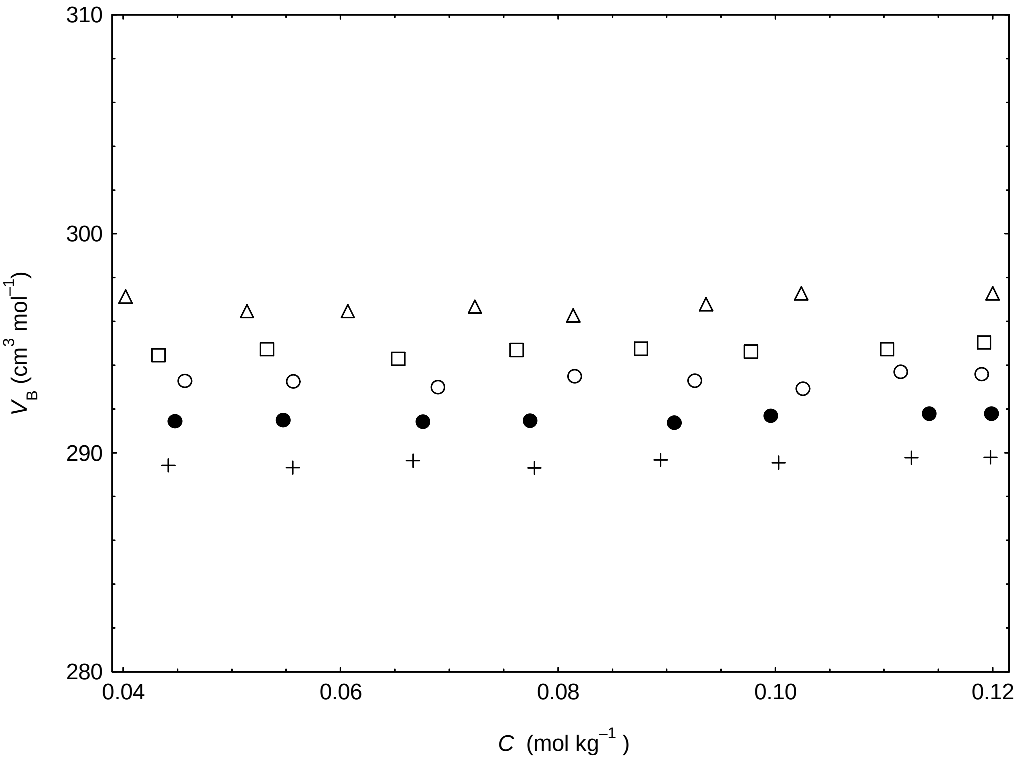

Figure 6. *Concentration independence of partial molar volume for dodecyltrimethylammonium bromide at 308.15 K, in pure water (triangles) and different methanol (1) + water (2) mixtures (squares, 0.10 methanol; circles, 0.20 methanol; closed circles, 0.30 methanol; crosses, 0.40 methanol).*

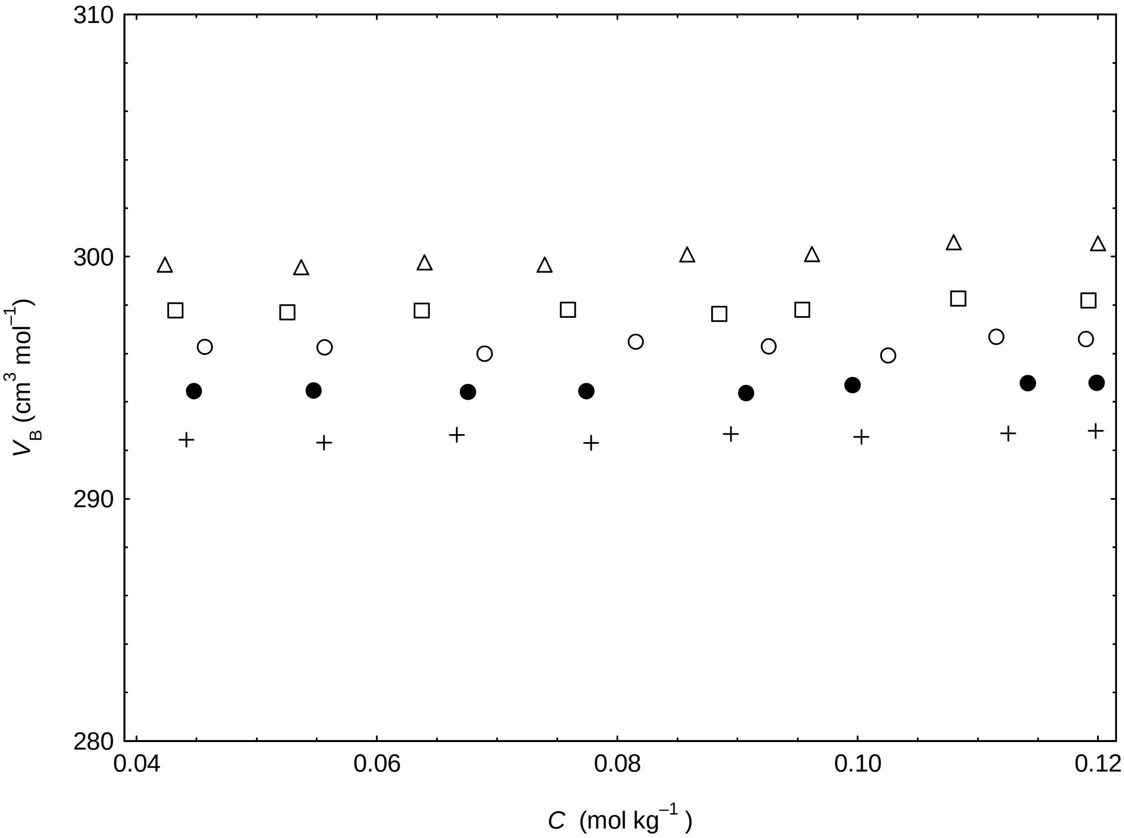

Figure 7. *Concentration independence of partial molar volume for dodecyltrimethylammonium bromide at 318.15 K, in pure water (triangles) and different methanol (1) + water (2) mixtures (squares, 0.10 methanol; circles, 0.20 methanol; closed circles, 0.30 methanol; crosses, 0.40 methanol).*

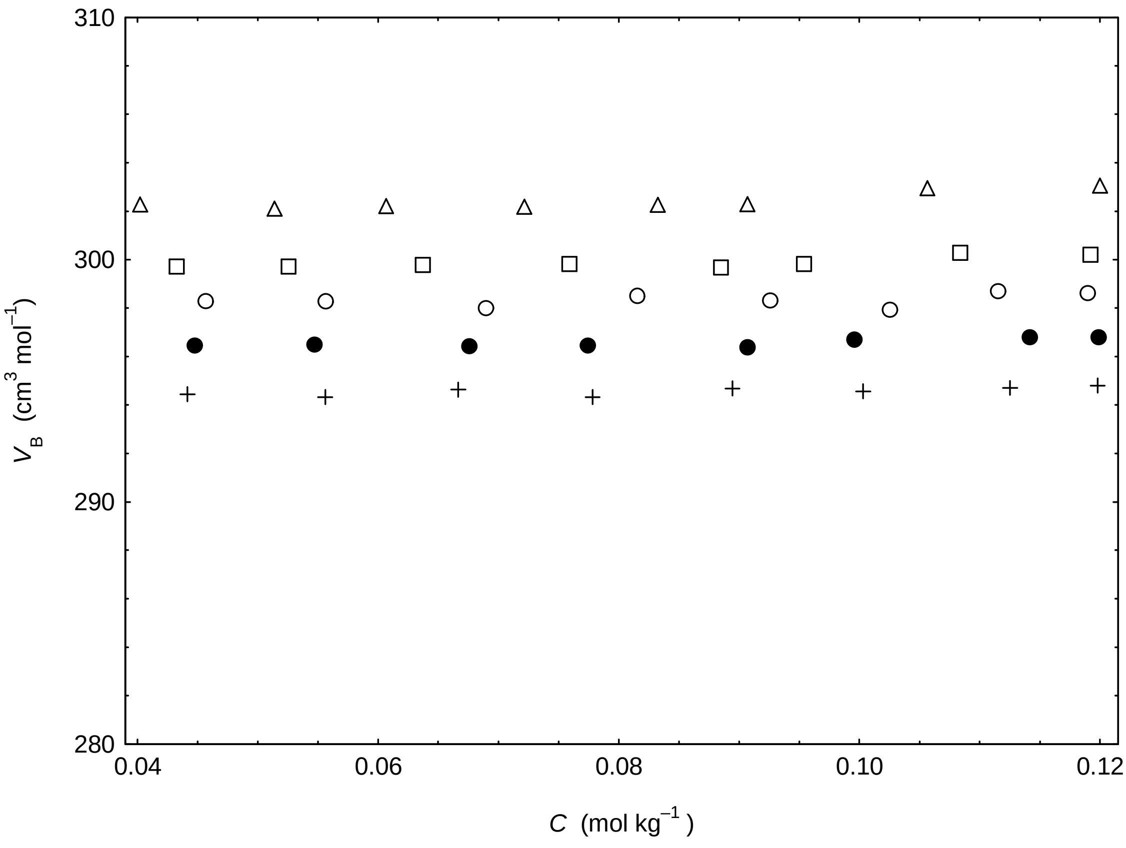

Figure 8. *Concentration independence of partial molar volume for dodecyltrimethylammonium bromide at 323.15 K, in pure water (triangles) and different methanol (1) + water (2) mixtures (squares, 0.10 methanol; circles, 0.20 methanol; closed circles, 0.30 methanol; crosses, 0.40 methanol).*

Our partial molar volume data for pure water of dodecyltrimethylammonium bromide match with the previously published papers(16, 23). Obviously, the concentration independence of partial molar volumes follows the same pattern at all the temperatures and solvent compositions investigated.

The effects of temperature and relative permittivity on the partial molar volume values have been shown in Table 2. At each temperature, the partial molar volume values are found to decrease with decreasing relative permittivity by increasing the methanol content in the system. On the other hand, the partial molar volume is increased in the given system with increasing temperature. This is mostly due to the weakening of surfactant-solvent binding energy with increasing temperature. The same pattern has been also reported in the work (24).

The relative permittivity of the medium is decreased with increasing in the methanol content at a given temperature and similar findings were reported in the previous works also (5, 13, 25, 26).

4. Conclusions

Experimental results for the density of salt-free solution of a cationic surfactant dodecyltrimethylammonium bromide in pure water and methanol-water mixed solvent media have been presented as a function of surfactant concentration and temperature. The densities are found to decrease with increasing temperature over the entire concentration range investigated in a given mixed solvent medium whereas these values are also found to decrease as the relative permittivity of the medium decreases. Estimation of the slopes and the calculated solvent density provide important insight regarding the solution behavior of cationic surfactant in methanol-water mixtures. With the help of density measurement, the calculated partial molar volumes of dodecyltrimethylammonium bromide have been presented as a function of surfactant concentration and temperature. The partial molar volumes are found to increase with increasing temperature over the entire concentration range investigated in a given mixed solvent medium. Furthermore, at a particular temperature, the partial molar volumes are found almost same even at the different concentrations and these values are found to be decreased as the relative permittivity of the medium decreases.

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