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Conductivity, Density, and Adiabatic Compressibility of Dodecyldimethylbenzylammonium Chloride in Aqueous Solutions

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Conductivities of dodecyldimethylbenzylammonium chloride in aqueous solutions were studied in the 15−35 °C temperature range at 5 °C intervals. Densities of the solutions were studied at 15, 20, and 25 °C. Adiabatic compressibility of the aqueous solutions of the surfactant has been determined from measurements of ultrasound velocity and density at 25 °C. The methods yielded identical critical micelle concentration (cmc) values. Conductivity results show a shallow minimum of the cmc values at around 25 °C. The ionization degree of micelles, β, increases, whereas the standard free energy of micellization, ΔG_{m}°, decreases upon growth of temperature. From density data for the surfactant, the change of the apparent molar volume upon micellization was calculated. The apparent adiabatic compressibility for the micelle of the surfactant at 25 °C was estimated. For comparative purposes we have measured additionally the conductivity of the decyl homologue at 30 °C to estimate the cmc and β values, and at 25 °C to estimate the value of the limiting equivalent conductivity.

Introduction

A survey of the literature on micellar properties of alkyldimethylbenzylammonium salts in aqueous solutions was presented by us in a recent paper in which we have reported results of our conductometric (at 15, 25, 35, 45, and 55 °C) and densitometric (at 15, 25, and 35 °C) studies on decyldimethylbenzylammonium chloride, (C10BCl).[1] We found that introduction of a phenyl group in place of a hydrogen atom at the *N*-trimethylammonium headgroup leads to a marked decrease of critical micelle concentration (cmc) value and an increase in the degree of ionization, β, of the micelles. The cmc values of the studied salt appear to be temperature-dependent and exhibit a shallow minimun around 35 °C. It was suggested that the change of apparent molar volume upon micellization of C10BCl is due mainly to the dehydration of the counterions bound to the micelles.

In this paper we present our results on conductometric and densitometric measurements at various temperatures and ultrasound velocity measurements, at 25 °C, of aqueous solutions of dodecyldimethylbenzylammonium chloride, (C12BCl). These are supplemented by some results of conductometric measurements of C10BCl at 25 and 30 °C.

Materials and Methods

The title compound, C12BCl, was synthesized by quaternization of *N*,*N*-benzyldimethylamine with dodecyl chloride by applying a procedure similar to that used for the synthesis of the decyl homologue.[1] The surfactant was purified by several recrystallizations from acetone. The air-dried product appeared to be a monohydrate as determined by the Karl Fischer method.
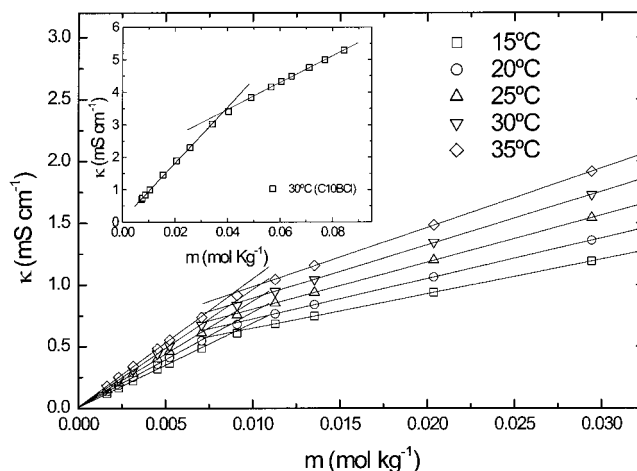
Figure 1. Dependence of specific conductivity of C12BCl on molality at various temperatures and of C10BCl at 30 °C (insert).

Electrospray mass spectroscopy data indicated that the salt was not contaminated by other homologues.

Conductivity and density measurements were carried out as described earlier.[1] Ultrasound velocity of aqueous solutions of C12BCl was measured at 25 ± 0.01 °C, at a frequency of 2 MHz using a Nusonic model 6380 concentration analyzer with an accuracy of ± 0.01m/s.

Results

The plots of specific conductivity versus molality for C12BCl at various temperatures and a plot for C10BCl at 30 °C are shown in Figure 1. They exhibit typical behavior with two linear fragments whose intersection is usually treated as corresponding to the cmc. It has been suggested that the ratio of the slopes of the linear fragments above and below cmc gives an estimate of β of the micelles.[2,3]
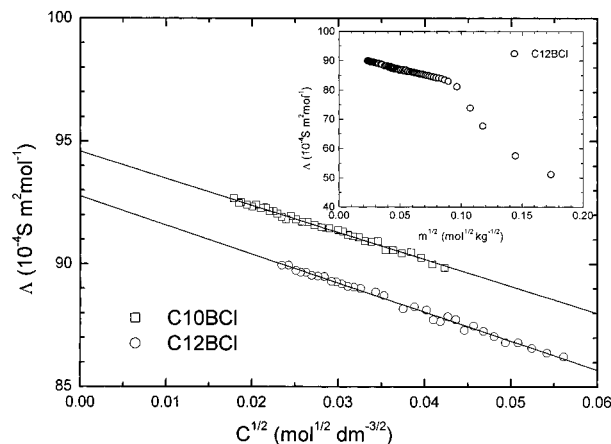
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TABLE 1: Values of m_{cmc}, β, and ΔG_m° for C12BCl at Various Temperatures and for C10BCl at 30 °C[a]

t(°C)	m_{cmc} (mol kg^{-1})	β	ΔG_m° (kJ mol^{-1})
15	0.00913	0.41	-33.1_5
20	0.00893	0.43	-33.5
25	0.00880	0.45	-33.7
30	0.00883	0.46	-34.0
35	0.00899	0.46_5	-34.3
30[a]	0.0379	0.49	-27.7

Figure 2. Dependence of equivalent conductivity on $C^{1/2}$ of C10BCl and C12BCl in the low concentration range at 25 °C. The plot for the whole concentration range of C12BCl is shown in the insert.

According to the charged phase separation model of micellization, the activity of the monomer remains constant above cmc,[4] and the standard free energy of micellization, ΔG_m°, can be calculated from the following relation:

$$\Delta G_m^\circ = (2 - \beta)RT \ln X_{cmc} \quad (1)$$

where X_{cmc} is expressed in mole fraction units. The temperature-dependent values of m_{cmc}, i.e., molalities at which an abrupt change of slope on the conductivity vs molality plots occur, as well as of β and of ΔG_m° are presented in Table 1. Included are the corresponding values for C10BCl at 30 °C.

Figure 2 shows values of equivalent conductivity, Λ, of C10BCl and of C12BCl at 25 °C, in the lowest concentration range, plotted in function of square root of concentration. In the insert is shown the plot for the whole concentration range studied for C12BCl at 25 °C.

The results of density, ρ, measurements at 15, 20, and 25 °C are presented in Figure 3 in the form of plots of $\rho - \rho_o$ versus molality of C12BCl, ρ_o being the density of water at the appropriate temperature. It is seen that in the whole postmicellar concentration range studied, the densities decrease linearly with increase of molality. In the premicellar range, the concentration dependence of density is also linear; however, as can be seen in the insert in Figure 3, the slopes change upon increase in temperature, passing gradually from positive to negative values. The results show that the change in sign of the slope occurs in the 15−20 °C temperature range.

The apparent molar volumes, V_ϕ, of C12BCl in function of molalities at the temperatures studied were calculated using the following equation:

$$V_\phi = \frac{M}{\rho} + \frac{10^3(\rho_o - \rho)}{m\rho_o\rho} \quad (2)$$

where M and m are the molecular mass and the molality of the
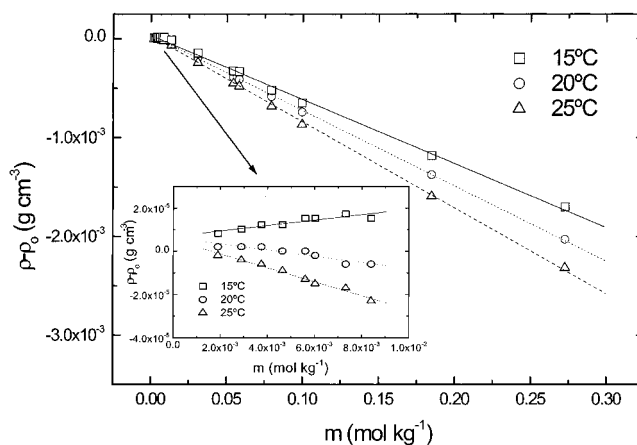
Figure 3. Density vs molality plot for C12BCl at 15, 20, and 25 °C. The plot for premicellar molality range is displayed in the insert.
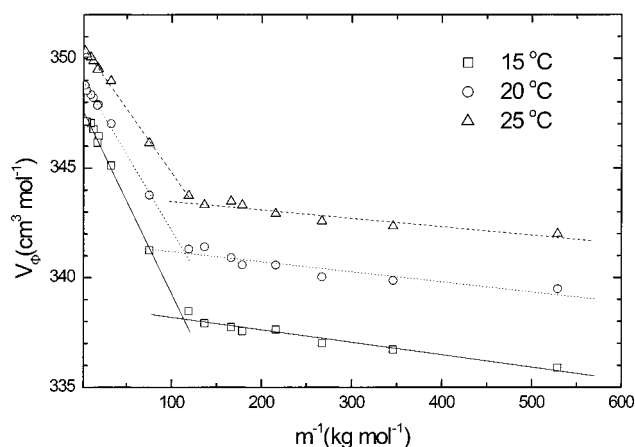
Figure 4. Variation of V_ϕ with the reverse of molality for C12BCl at 15, 20, and 25 °C.
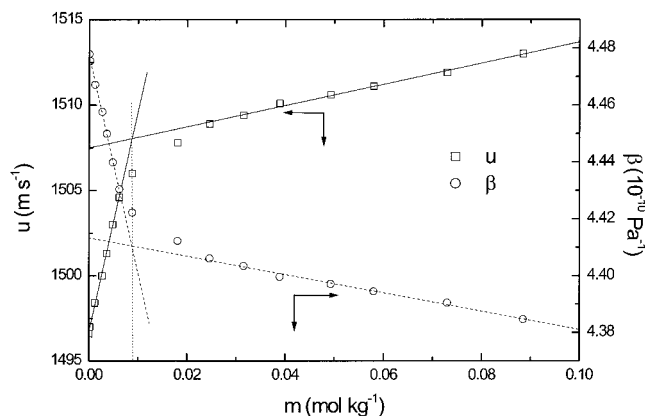
Figure 5. Effect of C12BCl molality on ultrasound velocity u and adiabatic compressibility β_s in aqueous solutions at 25 °C.

solute, respectively. The variation of V_ϕ with the reverse of molality is illustrated in Figure 4.

Figure 5 shows the dependence of ultrasound velocity, u, measured for aqueous solutions of C12BCl as a function of molality at 25 °C and the corresponding dependence of adiabatic compressibility, β_s, calculated from the relation $u^2 = 1/\rho\beta_s$, where u is expressed in m s^{-1} and ρ in kg m^{-3}. The vertical dotted line indicates the intersection of the linear pre- and postmicellar fragments of both dependencies. It corresponds exactly to the m_{cmc} value of 0.0088 mol kg^{-1} obtained from our conductivity measurements.
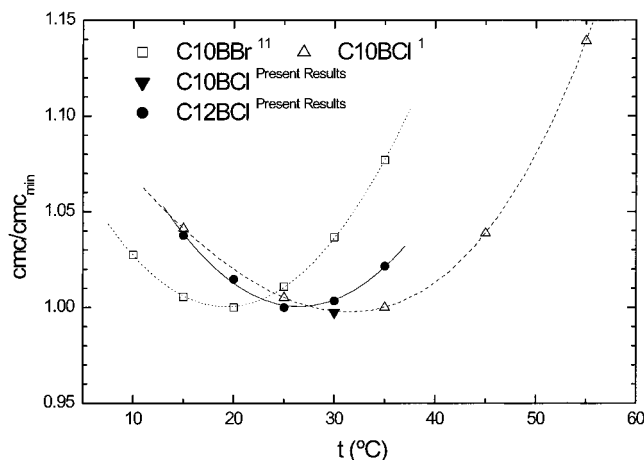
Figure 6. Temperature dependence of relative cmc values for C10BCl, C12BCl, and C10BBr.

Discussion

The cmc values determined conductometrically for C12BCl at 25 °C confirm earlier results.[5-8] Comparison of the cmc value for C12BCl at 25 °C with that determined by us recently for C10BCl (0.039 mol/kg)[1] shows that the value of cmc decreases by a factor of 2.105 with the increase of the length of the alkyl chain by one methylene group. This factor is comparable with those established for homologous alkyltrimethylammonium chlorides and alkylpyridinium chlorides[9] and for alkylpyridinium bromides[10] and shows that the rate of change in cmc per one methylene group in the long alkyl chain of cationics does not depend markedly on the structure of the headgroup and type of counterion.

The temperature dependence of cmc for C12BCl is illustrated in Figure 6 in the form of relative changes, where included are the corresponding values taken from the data of Adderson and Taylor for C12BBr[11] and the data for C10BCl[1] supplemented by the present result obtained at 30 °C. It can be seen that the minimum is shifted toward lower values of temperature upon passing to a more hydrophobic cation (C10BCl to C12BCl) as well as to a more hydrophobic anion (C12BCl to C12BBr). This phenomenon for the hydrophobic cations[12] and nonionic surfactants[13] has been already well documented.

The estimated values of degree of ionization, β, of the micelles of C12BCl shown in Table 1, increase with temperature as found earlier for C10BCl[1] and for alkyltrimethylammonium bromides.[14,15] The mean values of $\Delta\beta/\Delta T$ estimated in the temperature range of 288 to 308 K amount to 0.0030 and 0.0028 K^{-1} for C10BCl and C12BCl, respectively. These values are comparable to those found earlier for tetradecyltrimethylammonium bromide (0.003)[16] and for tetradecyldimethylphenylammonium bromide (0.0029).[17]

The standard free energy of micellization, calculated for C10BCl and C12BCl by applying eq 1, shows that at 25 °C the difference per one mole of methylene groups amounts to 2.95 kJ. ΔG_m° for C12BCl decreases linearly with growth of temperature and can be described by the relation: ΔG_m° (J mol^{-1}) $= -17034 \pm 688 - (58 \pm 2.3)T$, for T in the range of 288 to 308 K. The low rate of decrease of ΔG_m° with growth of temperature is typical for aqueous solutions of surfactants and results from an entropy−enthalpy compensation effect.[13,17-20]

The conductivity data, obtained in the 15−35 °C range, presented in the form of plots of equivalent conductivity versus square root of molality do not reveal anomalies in the vicinity of cmc as reported by Markina et al.[8] Such a presentation, for

25 °C, is exemplified in the insert of Figure 2. Our results do not confirm the occurrence of premicellar dimerization of the two homologues in the temperature range studied. The two straight lines shown in Figure 2 represent the linear fits to the experimental results for C10BCl and C12BCl at 25 °C, in the lowest concentration range available for our measurements. Extrapolation to zero concentration gives values of limiting equivalent conductivities uncorrected for activity, Λ_o. The values amount to 94.6 and 92.8 $\times 10^{-4}$ S cm^2 mol^{-1} for C10BCl and C12BCl, respectively. The difference of 0.9 $\times 10^{-4}$ S cm^2 mol^{-1} per one methylene group in the long chain is exactly the same as that found for the data reported for decyltrimethylammonium and dodecytrimethylammonium ions.[21] Note however, that the additivity rule for the values of limiting equivalent conductivities appears to be valid only when ionic amphiphiles with sufficiently long chains are being compared. In the vicinity of the ionic head, the methylene groups may be shielded to an extent depending on its bulkiness. This can be readily seen upon comparing the limiting equivalent conductivity data for the following two pairs of cations Λ_0^+: tetramethylammonium−N-dodecyltrimethylammonium $= 44.9 - 23.8 = 21.1$ and benzyltrimethylammonium−dodecyldimethylbenzylammonium $= 34.6 - 16.5 = 18.1 \times 10^{-4}$ S cm^2 mol^{-1}. The values of Λ_o for the first three cations were taken from ref 21 whereas the value for the C12B$^+$ cation results from subtraction of the value of Λ_o for the chloride anion,[21] amounting to 76.3, from the value of 92.8 $\times 10^{-4}$ S cm^2 mol^{-1} found presently for C12BCl. As expected, the bulky head of C12B$^+$ exerts a strong shielding effect equivalent to three methylene groups.

We will now discuss our density data. In an ionic surfactant system, below cmc, the dependence of the density on concentration can be expressed as

$$(\rho - \rho_o)10^3 = [(M_c + M_f) - (V_c + V_f)\rho_o]c \qquad (3)$$

where M_f, V_f, and M_c, V_c, are the molecular masses and the apparent molar volumes of the surfactant ions and counterions, respectively. If these apparent molar volumes were negligibly concentration-dependent, then eq 3 predicts a linear relation between density and concentration. Indeed, such a behavior is seen in the insert of Figure 4. Thus estimated values for $V_c + V_f$ amount to 338.6, 341.7, and 344.2 cm^3 mol^{-1} at 15, 20, and 25 °C, respectively. These values can be taken as zero-order approximation of the accurate values.

The change in the slope of the plots of density vs molality from positive to negative values, appearing in the insert of Figure 3, indicates, according to eq 3, that the $V_c + V_f$ term increases with temperature. This effect is related to the relaxation of structured water engaged in the solvation of the hydrocarbon chain, the headgroup, and the counterion upon increase in temperature. A similar behavior can be observed for the data for C10BCl.[1] The rate of change of the slope with temperature, $\Delta(\partial\rho/\partial c)/\Delta T$, is distinctly higher for C12BCl (-5.02×10^{-4} g mol^{-1} °C^{-1}) than for C10BCl (-3.15×10^{-4} g mol^{-1} °C^{-1}), as expected, and indicates the contribution of desolvation of the methylene groups in the hydrocarbon tail.

To obtain the value of the apparent molar volume at infinite dilution, V_ϕ^o, the values of V_ϕ obtained from eq 2 were fitted to the Debye−Hückel limiting law:

$$V_\phi = V_\phi^o + A_v m^{1/2} + B_v m \qquad (4)$$

where A_v is the Debye−Hückel coefficient which amounts to 1.696, 1.781, and 1.865 cm^3 kg$^{1/2}$ mol$^{-3/2}$ at 15, 20, and 25 °C, respectively, and B_v is an adjustable parameter which measures

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TABLE 2: Apparent Molar Volumes, of the Surfactant at Infinite Dilution, V_ϕ^o, at cmc, V_ϕ^{cmc}, and the Change upon Micellization, ΔV_ϕ^{cmc}, for C12BCl at 15, 20, and 25 °C

T (°C)	V_ϕ^o (cm³ mol⁻¹)	V_ϕ^{cmc} (cm³ mol⁻¹)	ΔV_ϕ^{cmc} (cm³ mol⁻¹)
15	335.6	338.4	9.35
20	338.9	341.4	7.74
25	341.6	344.0	6.60

deviations from the limiting law. The calculated values for V_ϕ^o are shown in Table 2.

Comparison of the values of V_ϕ^o for C12BCl and C10BCl[1] at 15 and 25 °C shows that the incremental differences per methylene group amount to 14.2 and 15.45 cm³ mol⁻¹, respectively. The former value seems to be low; note, however, that a much lower value, amounting to 10.0[22] cm³ mol⁻¹ can be extracted from the data reported for the corresponding homologues of alkyltrimethylammonium bromides at 25 °C. The V_ϕ^o values for C12BCl and C10BCl[1] increase almost linearly with temperature in conformity with observations for other surfactant systems.[23]

Assuming that the rule of additivity for the apparent molar volumes of the ions in the system is valid, we can write

$$n_{C12BCl}V_\phi = n_f V_f + n_c V_c + n_m V_m \qquad (5)$$

where V_m is the apparent molar volume of the micelles in the system and n_{C12BCl} the total number of moles of the surfactant, whereas n_f, n_c, and n_m are the numbers of moles of free monomers, free counterions, and of micelles, respectively.

Since the studied salt is a 1:1 electrolyte, then, according to the pseudo-phase separation model we have below cmc:

$$m_f = m_c = m \qquad (6a)$$

and above cmc:

$$m_f = cmc$$

$$m_c = cmc + \beta(m - cmc) \qquad (6b)$$

$$m_m = \frac{m - cmc}{N_{agg}}$$

where N_{agg} is the aggregation number of the micelle.

Dividing eq 5 by the mass of the solvent (in kg) and taking into account relations in eqs 6, we obtain the following set of equations:

$$V_\phi = (V_f + V_c) \quad m \leq cmc \qquad (7a)$$

$$V_\phi = \left(\frac{V_m}{N_{agg}} + \beta V_c\right) - \left[\frac{V_m}{N_{agg}} + (\beta - 1)V_c - V_f\right]\frac{cmc}{m}$$
$$m \geq cmc \quad (7b)$$

If, as previously, we assume that the molar volumes of the ionic species are not concentration-dependent, then, from the last equation a linear decrease of the V_ϕ vs $1/m$ plot can be expected in the concentration range above cmc. According to eq 7a the value of V_ϕ should be constant in the concentration range below cmc. Such a plot is shown in Figure 4. It can be seen that the system conforms approximately to the expected behavior. In the region where $(cmc/m) \geq 1$, a slight decrease of $V_f + V_c$ with m⁻¹ is, however, observed probably due to the

neglect of activity effects in the equations. Nevertheless, eqs 7 may be considered as an acceptable approximation of the behavior of the system.

Linear fitting of the points in the $cmc/m \leq 1$ region allows us to obtain the value of the apparent molar volume, V_ϕ^{cmc}, (intercept + slope), of the surfactant at cmc, and the change in the apparent molar volume upon micellization, ΔV_ϕ^{cmc} (slope). The results are collected in Table 2.

Comparison of the values of V_ϕ^{cmc} obtained by applying eq 7b (for data above cmc) with the corresponding values of $V_f + V_c$ obtained via eq 3 (for data below cmc) shows deviations of about 0.2 to 0.3 cm³ mol⁻¹ which can be taken as a measure of the errors in the results.

The values of ΔV_ϕ^{cmc} found for C12BCl are positive. A similar observation was reported earlier for other surfactants.[1,24-27] It is generally assumed that this effect is mainly due to the release of structured water in the hydration shell of the monomers when the micelles are formed.

According to eq 7b, the slope of the V_ϕ vs cmc/m plot, ΔV_ϕ^{cmc}, shows a positive dependence on the degree of ionization. This last magnitude, as was pointed out above, grows with temperature. Thus, an increase in the Δ_ϕ^{cmc} values should be expected when the temperature is raised. This effect might be interpreted as being due to growth in the electrostatic repulsion between the ionic headgroups at the surface of the micelles. Results in Table 2 show, however, a reverse behavior, i.e., ΔV_ϕ^{cmc} decreases with growth of temperature. Again, the structured water surrounding the hydrocarbon chain of monomers is the origin of this phenomenon. At higher temperatures, the water sheath is less structured, and thus the contribution to ΔV_ϕ^{cmc} due to the release of water molecules is much lower. This effect is large enough to compensate the growth in ΔV_ϕ^{cmc} due to the increase of the electrostatic repulsion that occurs when the degree of ionization rises.

Our results for ΔV_ϕ^{cmc} found for the two studied homologues are in conformity with the above given discussion. And thus, the ΔV_ϕ^{cmc} values for the higher homologue are larger by an amount of 2.3 and 0.9 cm³ mol⁻¹ at 15 and 25 °C, respectively, which correspond to values of 1.15 and 0.45 cm³ mol⁻¹ per one methylene group. It is worth noting that for nonyl- and decyltrimethylammonium bromides, De Lisi et al.[24] reported values of 1.15 and 0.59 cm³ mol⁻¹ at 15 and 25 °C, whereas the data of Causi et al.[25] for decyl- and dodecylpyridinium chlorides at 25 °C yield a value of 0.75 cm³ mol⁻¹ per one methylene group.

For surfactant systems of sufficiently low concentration, the dependence of the adiabatic compressibility, β_S, on molality, (below and above cmc), can be expressed by the two corresponding equations given below. They may be treated as approximations of the exact equations used by Zielinski et al.:[26]

$$\frac{1000(\beta_S - \beta_{S_o})}{\rho_o} = (\bar\beta_{S_1} - \beta_{S_o})V_{\phi 1}m \quad m < cmc \qquad (8a)$$

$$\frac{1000(\beta_S - \beta_{S_o})}{\rho_o} = (\bar\beta_{S_m} - \beta_{S_o})V_{\phi 1}(m - cmc) \quad m > cmc$$
$$(8b)$$

where $V_{\phi 1}$ and $V_{\phi m}$ are the apparent molar volumes of the system below and above the cmc, ρ_o is the density of pure water (in g cm⁻³), and β_{S_o}, $\bar\beta_{S_1}$, and $\bar\beta_{S_m}$ are the adiabatic compressibility of pure water and the apparent adiabatic compressibilities of
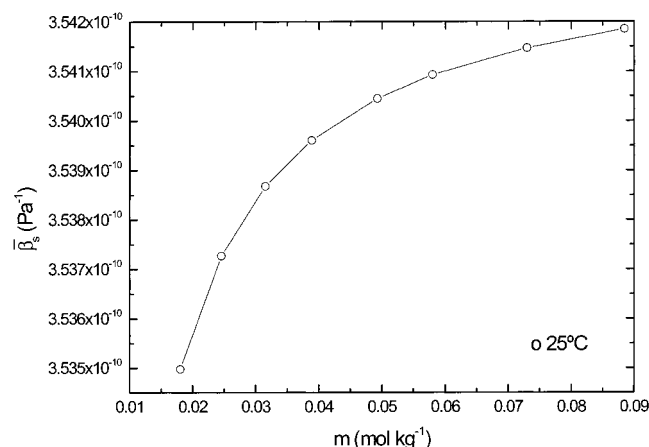
Figure 7. Molality dependence of the apparent adiabatic compressibility of C12BCl at 25 °C.

surfactant in the monomeric and micellar states, defined respectively by

$$\beta_{S_o} = -\frac{1}{V_o}\left(\frac{\partial V_o}{\partial P}\right)_S, \quad \bar{\beta}_{S_1} = -\frac{1}{V_{\phi_1}}\left(\frac{\partial V_{\phi_1}}{\partial P}\right)_S, \quad \bar{\beta}_{S_m} = -\frac{1}{V_{\phi_m}}\left(\frac{\partial V_{\phi_m}}{\partial P}\right)_S \quad (9)$$

Figure 5 shows that the dependence of β_s on molality is well represented by two straight lines of different slopes in the pre- and post-micellar concentration range. From the value of the slope above cmc, the value of $\bar{\beta}_{S_m}$ can be estimated by applying eq 8b. The results obtained are shown in Figure 7. The value of $\bar{\beta}_{S_m}$ increases asymptotically to a limiting value of 3.544×10^{-10} Pa^{-1} which compares well with the corresponding data reported by several authors for homologues alkyltrimethylammonium bromides. [26,28,29]

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