Micellisation thermodynamics of sodium lauroylsarcosinate in water–alcohol binary mixtures

Olanrewaju Owoyomi*, Oludare Alo, Oladega Soriyan and Grace Ogunlusi

Department of Chemistry, Obafemi Awolowo University, Ile Ife, Nigeria

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Aggregation of sodium lauroylsarcosinate (SLS) in aqueous solutions of methanol, ethanol, propanol and ethylene glycol at 288–313 K has been determined from conductivity measurement in the range 0–20% v/v of additives. The precise values of the critical micelle concentration (CMC) and the degree of counter-ion dissociation of micelles were obtained at each temperature by fitting the specific conductivity-surfactant concentration curve to the integrated form of the Boltzmann-sigmoid equation. The CMC was found to increase with increase in additive concentrations in the case of methanol and ethylene glycol, while it decreases with increase in ethanol and propanol concentration. The equilibrium model of micelle formation was applied to obtain the thermodynamic parameters of micellisation. The Gibbs free energies were observed to vary only slightly with temperature and additive concentrations. Enthalpy–entropy compensation was observed for all the systems with a constant compensation temperature of ≈300 K and negative compensation enthalpy.

Keywords: alcohols; sodium lauroylsarcosinate; micellisation; thermodynamic parameters; enthalpy–entropy compensation

1. Introduction

The formation of surfactant aggregates in mixed aqueous-organic solvents systems has been extensively studied with the aim of characterising the effects of the solvent properties on the aggregation behaviour [1–6]. Polar organic solvents with properties similar to those of water, such as dimethylsulphoxide, acetonitrile, dimethylformamide, ethylene glycol, glycerol and monohydric alcohols, which are characterised by high dielectric constant, high cohesive energy and considerable hydrogen bond, have been widely studied [7–15]. The alcohols apparently are the most widely studied additives probably because of their important role in the preparation of microemulsions [16–20].

It is generally believed that the alcohol is partitioned between the water and the micellar pseudo-phase with the alcohol molecules in the micellar pseudo-phase intercalated between the surfactant head groups by replacing the water molecules around the ionic head groups, thus increasing the electrostatic repulsion between the surfactant head groups [21,22]. This also leads to a decrease in the dielectric constant at the micellar surface [23].

Sodium lauroylsarcosinate (SLS) is one of the salts of N-acylsarcosine, a group of amino-acid-based surfactants, which is gaining importance in industrial as well as personal care and cosmetic applications because it has been found to be mild, less irritating to

*Corresponding author. Email: oowoyomi@oauife.edu.ng

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the skin, easily biodegradable, has better stability towards hard water and possesses antimicrobial activities [24, 25].

Although a considerable insight has been gained about the factors governing the surface and bulk properties of aqueous solutions of SLS, such as the effect of temperature, added electrolyte and pH variation on its aggregation behaviour [26], no work has been reported on its micellar properties in the presence of organic co-solvent; hence, it follows that the solution and interfacial properties of this sarcosinate have not been adequately studied.

Moreover, most practical applications of surfactants involve the presence of other species, such as glycols and monohydric alcohols; it is therefore important to establish the effects of such and related compounds on the micellisation process in order to explore their fundamental behaviour. In view of its immense biological and industrial importance, such information is essential for its profitable applications and uses in practice. In this work, we have investigated the effects of chain length as well as the number of hydroxyl group of alcohols on the micellisation of SLS.

2. Experimental

2.1. Chemicals

The surfactant SLS was purchased from Sigma Aldrich with ≥94% purity and was used without further purification. Analytical grade methanol, ethanol and 1-propanol were also purchased from Sigma Aldrich and were also used without further purification. Ethylene glycol (99% purity) was from British Drug House (BDH) and was re-distilled to ascertain its purity.

2.2. Method

All solutions were prepared using glass-distilled water with conductivity not greater than 3 μS·cm⁻¹ at 25°C. The conductivity measurements were made with CMD 210 digital conductivity meter from Walden Precision Apparatus, United Kingdom. The cell constant was determined by calibration with several standard solutions of KCl of known specific conductivities. Conductometric titration method, involving the titration of a known volume of surfactants into a fixed volume of water contained in a thermostatted beaker, was employed. All measurements were made in a thermostatted water bath (Grant Y14, Grant Instruments Ltd., Cambridge, UK), maintaining the temperature constant within ±0.1°C.

3. Results and discussion

3.1. Determination of critical micelle concentration

The conductivity of increasing concentrations of SLS in different water–co-solvent mixtures was measured at \( T = 288, 293, 298, 303, 308 \) and 313 K. The conductivity can be linearly correlated to the surfactant concentration in both the pre-micellar and the post-micellar regions. The intersection point between the two straight lines gave the critical micelle concentration (CMC). The slope in the pre-micellar region is greater than in the post-micellar region and the ratio of the slopes of the post-micellar region to that of the pre-micellar region gives the effective degree of counter-ion dissociation. However, it was found that an increase in the percentage by volume of the alcohols results in a less abrupt change in conductivity in going from the surfactant concentration
in the pre-micellar range to surfactant concentration in the post-micellar range, as compared to that in pure water, thus making the determination of the CMC values difficult with an increase in the percentage by volume of the alcohols. To overcome this, the method proposed by Carpena et al. [27] was adopted for the accurate determination of the CMC and the degree of counter-ion dissociation values that are necessary for calculating the thermodynamic parameters.

The method involves fitting of the raw experimental conductivity data, \( \kappa \), as a function of surfactant concentration, \( c \), to the equation

\[
\kappa(c) = \kappa(c = 0) + A_1 c + \Delta c (A_2 - A_1) \ln \left[ \frac{1 + e^{(c-c_0)/\Delta c}}{1 + e^{-(c-c_0)/\Delta c}} \right]
\]  (1)

Here, \( \kappa(c = 0) \) represents the conductivity of solution when \( c = 0 \), \( A_1 \) and \( A_2 \) represent the pre-micellar and post-micellar slopes, \( \Delta c \), the width of transition whose central point, \( c_0 \), corresponds to the CMC. Plots of conductivity against SLS concentration are shown in Figures 1 and 2. The CMC, \( A_1, A_2 \) and the degree of counter-ion dissociation values (\( \alpha = A_2/A_1 \)), obtained directly from the fittings of conductivity–concentration plots to Equation (1) are summarised in Tables 1 and 2.

At a given temperature, the CMC of SLS increases with an increase in concentration of methanol and ethylene glycol through all the additive concentrations studied whereas, the CMC values decrease from 5 to 15% v/v of ethanol before increasing again at 20% v/v of ethanol. The CMC values decrease with an increase in propanol concentration up to

![Figure 1. Plot of specific conductivity vs. [SLS] in water (open circle), 10% v/v methanol (open inverted triangle) and 10% v/v ethanol (cross).](image_url)
15% v/v of propanol beyond which no micellisation was observed. These variations in CMC values with an increase in co-solvent concentrations are shown in Figure 3. The observations above can be rationalised as follows; all the alcohols used as co-solvents in this study have significantly lower dielectric constant than water and consequently are expected to decrease the polarity of the medium and as a result decreasing the hydrophobic interactions between the hydrocarbon tails of the surfactant and hence delay the micellisation process to a higher concentration [28]. However, a second factor which favours the micellisation process is the possibility of the alcohol to penetrate the micellar core, intercalating between the surfactant head group thereby reducing the charge density at the micellar surface with a concomitant decrease in the electrostatic repulsion between the ionic head groups of the surfactant; this effect will increase with an increase in chain length of the alcohol [28,29].

In water–methanol systems, the former factor appears to predominate and this resulted in an increase in CMC values with an increase in methanol concentration. The increase in CMC with an increase in ethylene glycol concentration in the ethylene glycol–water mixtures was also due to the first factor explained above, with a slight contribution from the second factor because of the number of carbon atoms in ethylene glycol compared to methanol. At any given concentration, the increase in CMC values is greater in methanol than in ethylene glycol. For both ethanol and propanol, the second factor appears to predominate, hence the observed decrease in CMC values with an increase in concentration of either of these two co-solvents up to 15% v/v with the lowering in CMC values being higher in propanol than in ethanol.
Table 1. Values of the pre-micellar slope $A_1$ and post-micellar slope $A_2$ for the micellisation of SLS in different (water + co-solvent) mixtures of co-solvent volume per cent at various temperatures $T$ as obtained from Equation (1).

| Temperature / K | 288 | 293 | 298 | 303 | 308 | 313 |
|-----------------|-----|-----|-----|-----|-----|-----|
| Water           | 71316.42 | 34685.71 | 70056.58 | 36901.18 | 71358.44 | 37074.49 |
| Methanol % v/v  |     |     |     |     |     |     |
| 5               | 60878.73 | 32131.65 | 60800.03 | 33428.55 | 62691.86 | 34918.53 |
| 10              | 55822.58 | 30193.17 | 58709.25 | 31048.47 | 60131.89 | 34810.97 |
| 15              | 52749.31 | 25714.62 | 52278.52 | 31637.39 | 53750.32 | 35502.22 |
| 20              | 47610.51 | 31378.09 | 51023.65 | 30848.34 | 52958.39 | 32309.97 |
| Ethanol % v/v   |     |     |     |     |     |     |
| 5               | 62170.89 | 32741.56 | 64382.59 | 33440.48 | 68100.72 | 33138.97 |
| 10              | 52946.48 | 30779.15 | 54837.32 | 35053.55 | 59898.92 | 36602.62 |
| 15              | 49805.48 | 31243.97 | 52471.87 | 34561.62 | 57829.72 | 36469.63 |
| 20              | 42080.19 | 36045.62 | 49138.03 | 32351.62 | 51318.39 | 36493.06 |
| Propanol % v/v  |     |     |     |     |     |     |
| 5               | 60115.23 | 39412.49 | 60947.69 | 40785.54 | 64936.67 | 42412.01 |
| 10              | 49269.55 | 41703.63 | 54893.76 | 44146.43 | 55965.42 | 49280.49 |
| 15              | 51153.31 | 43478.31 | 51177.71 | 44526.28 | 50686.83 | 45962.30 |
| Ethylene glycol % v/v |     |     |     |     |     |     |
| 5               | 59164.21 | 30415.95 | 60567.00 | 32653.50 | 61868.79 | 33193.15 |
| 10              | 52209.29 | 29005.31 | 57078.84 | 29513.16 | 57298.02 | 30898.23 |
| 15              | 46877.67 | 25921.11 | 48421.50 | 26572.15 | 51081.43 | 26977.72 |
| 20              | 43279.62 | 23429.59 | 44099.72 | 23867.23 | 44977.50 | 24746.96 |

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Table 2. Calculated CMC values and degree of counter-ion dissociation \( \alpha \) for SLS in different (water + co-solvent) mixtures of co-solvent volume per cent at various temperatures \( T \).

| \( T/K \) | Water | Methanol (%v/v) | Ethanol (%v/v) | Propanol (%v/v) | Ethylene glycol (% v/v) |
|----------|-------|-----------------|----------------|-----------------|------------------------|
|          | [CMC] mol·dm\(^{-3}\) | \( \alpha \) | [CMC] mol·dm\(^{-3}\) | \( \alpha \) | [CMC] mol·dm\(^{-3}\) | \( \alpha \) | [CMC] mol·dm\(^{-3}\) | \( \alpha \) | [CMC] mol·dm\(^{-3}\) | \( \alpha \) |
| 288      | 0.0132 | 0.49 | 0.0129 | 0.53 | 0.0125 | 0.52 | 0.0124 | 0.53 | 0.0126 | 0.54 | 0.0130 | 0.40 |
| 293      | 0.0142 | 0.53 | 0.0139 | 0.55 | 0.0136 | 0.56 | 0.0135 | 0.57 | 0.0139 | 0.58 | 0.0144 | 0.57 |
| 298      | 0.0144 | 0.54 | 0.0142 | 0.53 | 0.0140 | 0.58 | 0.0139 | 0.56 | 0.0143 | 0.63 | 0.0150 | 0.52 |
| 303      | 0.0154 | 0.49 | 0.0152 | 0.56 | 0.0145 | 0.66 | 0.0155 | 0.63 | 0.0146 | 0.64 | 0.0174 | 0.60 |
| 308      | 0.0158 | 0.66 | 0.0156 | 0.46 | 0.0154 | 0.61 | 0.0171 | 0.64 | 0.0190 | 0.63 | 0.0212 | 0.68 |
| 313      | 0.0142 | 0.53 | 0.0119 | 0.52 | 0.0117 | 0.49 | 0.0121 | 0.50 | 0.0123 | 0.45 | 0.0127 | 0.55 |
| 5        | 0.0118 | 0.58 | 0.0112 | 0.64 | 0.00107 | 0.65 | 0.0113 | 0.62 | 0.0120 | 0.64 | 0.0124 | 0.61 |
| 10       | 0.0104 | 0.63 | 0.0097 | 0.66 | 0.0089 | 0.63 | 0.0099 | 0.69 | 0.0107 | 0.66 | 0.0118 | 0.66 |
| 15       | 0.0122 | 0.86 | 0.0117 | 0.66 | 0.0108 | 0.71 | 0.0112 | 0.84 | 0.0119 | 0.84 | 0.0128 | 0.87 |
| 20       | 0.0105 | 0.66 | 0.0094 | 0.67 | 0.0087 | 0.65 | 0.0080 | 0.72 | 0.0091 | 0.66 | 0.0097 | 0.71 |
| 5        | 0.0099 | 0.84 | 0.0090 | 0.80 | 0.0079 | 0.88 | 0.0076 | 0.83 | 0.0085 | 0.86 | 0.0091 | 0.82 |
| 10       | 0.0088 | 0.85 | 0.0079 | 0.87 | 0.0069 | 0.91 | 0.0072 | 0.82 | 0.0082 | 0.76 | 0.0088 | 0.92 |
| 15       | 0.0135 | 0.51 | 0.0133 | 0.54 | 0.0130 | 0.54 | 0.0132 | 0.55 | 0.0134 | 0.55 | 0.0136 | 0.58 |
| 20       | 0.0137 | 0.56 | 0.0135 | 0.52 | 0.0131 | 0.54 | 0.0137 | 0.54 | 0.0142 | 0.54 | 0.0146 | 0.60 |
| 5        | 0.0139 | 0.55 | 0.0142 | 0.55 | 0.0144 | 0.53 | 0.0152 | 0.52 | 0.0158 | 0.59 | 0.0167 | 0.59 |
| 10       | 0.0142 | 0.54 | 0.0147 | 0.54 | 0.0152 | 0.55 | 0.0160 | 0.55 | 0.0169 | 0.60 | 0.0178 | 0.61 |
3.2. Thermodynamics of micellisation

The CMC values of the surfactant in the absence and presence of co-solvents showed a minimum in the plots of the variation of CMC with temperature (Figure 4). The minimum in these curves is noticed between 298 K and 303 K, which appears to conform to the general finding that the minimum for ionic surfactants with 12 carbon chain lengths is close to room temperature [30,31].

The relevant thermodynamic parameters, such as $\Delta G^o_M$, $\Delta H^o_M$ and $\Delta S^o_M$, of micellisation were determined on the basis of the phase separation model [32]. Accordingly

$$\Delta G^o_M = (2 - \alpha)RT \ln \chi_{\text{CMC}}$$

(2)

Figure 3. Plot of CMC vs. co-solvent concentrations at 298 K: methanol (open circle), ethylene glycol (open inverted triangle), ethanol (filled circle) and propanol (filled triangle).

Figure 4. Plot of CMC vs. temperature for the micellisation of SLS in water–co-solvent systems at 10% co-solvents: methanol (open circle), ethylene glycol (open inverted triangle), ethanol (filled circle) and propanol (filled triangle).
where $\chi_{CMC}$ is the value of CMC expressed on a mole fraction basis. The values of the degree of counter-ion dissociation ($\alpha$) did not change appreciably with temperature, hence, the average values of $\alpha$ were used in computing the thermodynamic parameters. The enthalpy of micellisation can be obtained from the temperature dependence of the CMC using the Gibbs–Helmholtz Equation (3).

$$\Delta H^o_M = -T^2 \frac{\partial \left( \frac{\Delta G^o_M}{T} \right)_P}{\partial T} = -RT^2 \left[ (2 - \alpha) \left( \frac{\partial \ln \chi_{CMC}}{\partial T} \right)_P \right]$$ (3)

In order to compute $\Delta H^o_M$, the variation of $\ln \chi_{CMC}$ with temperature was fitted to Equation (4), as proposed by Kim and Lim [33];

$$\ln \chi_{CMC} = A_o + A_1 \ln(T) + \frac{A_2}{T}$$ (4)

The entropy of micellisation can then be calculated from the values of the free energy and enthalpy obtained using Equations (2) and (3), respectively, as

$$\Delta S^o_M = \frac{\Delta H^o_M - \Delta G^o_M}{T}$$ (5)

The effect of the co-solvent on the micellisation process can be determined from the free energy of transfer defined by Equation (6) [34] as

$$\Delta G_{transfer} = \left( \Delta G^o_M \right)_{co-solvent/water} - \left( \Delta G^o_M \right)_{water}$$ (6)

The thermodynamic parameters calculated for the micellisation of SLS in different water–co-solvent mixtures at 298 K are listed in Table 3. Data for other temperatures are listed in Tables S1–S6 of the supplementary material.

The data given in Table 3 show that the free energy of micellisation becomes less negative with an increase in co-solvent concentration, indicating reduced spontaneity of the micellisation process in the presence of the co-solvents. The values for the enthalpy change of micellisation are, in most cases, positive at low concentration of the alcohols in the solvent mixtures and become increasingly less positive or more negative with an increase in the co-solvent concentration. The values of the standard entropy of micellisation of the surfactant are generally positive and become less positive, as the concentration of the co-solvents increases. The positive values of $\Delta S^o_M$ have been attributed to the disruption of water structure around the hydrocarbon tails of the surfactant monomers and the increased randomness of the hydrocarbon chains in the micellar core [9,35]. However, the micellisation process becomes entropy-destabilised with an increase in the concentration of the alcohol co-solvents, the $\Delta S^o_M$ values being less positive in all cases, this is due to the loss of water structuring that occurs with an increase in co-solvent concentrations; meanwhile, the values of $\Delta H^o_M$ become increasingly less positive, indicating that that at higher co-solvent concentrations, the contribution of the $\Delta H^o_M$ to the micellisation process becomes significant. The above observation could suggest that the micellisation process is entropy-driven at low concentrations of the co-solvents, but enthalpy-driven at high concentrations of the co-solvents.

The values of $\Delta G_{transfer}$ show that the effect of the co-solvent on the micellisation process is positive for all the co-solvents investigated with its value increasing with an
increase in the volume fraction of the co-solvents in the mixture. This results from the interaction of the hydrocarbon part of the surfactant with the hydrocarbon part of the co-solvents, which favours micellisation in the case of ethanol and propanol and increasing solubility of the hydrocarbon chain of the surfactant monomers, which disfavours micellisation as it is the case with methanol and ethylene glycol.

Table 3 also shows that the loss in entropic contribution to the micellisation process with an increase in co-solvent concentration is partially compensated for by the enthalpic gain. This is the so-called enthalpy–entropy compensation effect, which has been observed in a variety of processes including micellisation [36–38]. This phenomenon shows a linear correlation between the enthalpy and the entropy change and can be represented by the following equation

$$\Delta H_M^o = \Delta H^o + T_c \Delta S_M^o$$  \hspace{1cm} (7)

where $T_c$, the compensation temperature is the slope and is the intercept of the enthalpy–entropy compensation plot; such a typical plot is shown in Figure 5 below.

Generally, for all the systems investigated, the compensation plots are linear with correlation coefficient close to unity. The coefficients of the equation describing such plots are listed in Table 4.

The slope of these plots yields the compensation temperature $T_c$, which is often used to characterise solute–solute and solute–solvent interactions [37,38]; while the intercept of the plots gives the compensation enthalpy $\Delta H^o$, considers as the index of the chemical part of micellisation (solute–solute interactions). It stands for the enthalpy effect in the absence of any entropic contributions (i.e. at $\Delta S_M^o = 0$). The values of $T_c$ obtained in this work, both in

| % v/v co-solvent | $\Delta G_M^o$ (kJ·mol$^{-1}$) | $\Delta H_M^o$ (kJ·mol$^{-1}$) | $\Delta S_M^o$ (J·mol$^{-1}$·K$^{-1}$) | $\Delta G_{transfer}$ (kJ·mol$^{-1}$) |
|------------------|-------------------------------|------------------------------|-------------------------------------|-------------------------------|
| Water            | -31.26                        | 2.67                         | 113.88                              | —                             |
| Methanol         |                               |                              |                                     |                               |
| 5                | -29.58                        | 1.40                         | 103.95                              | 1.68                          |
| 10               | -29.36                        | 0.58                         | 100.48                              | 1.90                          |
| 15               | -28.25                        | -2.93                        | 84.97                               | 3.01                          |
| 20               | -27.26                        | -8.83                        | 61.85                               | 4.00                          |
| Ethanol          |                               |                              |                                     |                               |
| 5                | -31.22                        | 0.26                         | 105.63                              | 0.04                          |
| 10               | -28.91                        | -0.03                        | 96.92                               | 2.35                          |
| 15               | -28.80                        | -1.08                        | 93.03                               | 2.46                          |
| 20               | -25.07                        | 1.87                         | 90.39                               | 6.19                          |
| Propanol (% v/v) |                               |                              |                                     |                               |
| 5                | -28.60                        | 8.39                         | 124.12                              | 2.66                          |
| 10               | -25.20                        | 7.95                         | 111.25                              | 6.06                          |
| 15               | -25.08                        | 4.73                         | 100.02                              | 6.18                          |
| Ethylene glycol (% v/v) |   |                              |                                     |                               |
| 5                | -30.05                        | 0.72                         | 103.26                              | 1.21                          |
| 10               | -29.76                        | -1.45                        | 95.01                               | 1.50                          |
| 15               | -29.15                        | -6.81                        | 74.96                               | 2.11                          |
| 20               | -28.64                        | -8.99                        | 65.95                               | 2.62                          |
the absence and presence of the additives are within the range $300 \pm 3$ K, which implies that the presence of the co-solvents has little or no effect on the solute–solvent interactions. The values of the compensation temperature fits well in the general framework proposed by Sugihara and Hisatomi, in which all surfactants should be included in the range from 299 to 315 K [38]. The values of $\Delta H^*$, the intrinsic enthalpy gain are all negative indicating that the micellisation process is favoured even in the absence of any entropic gain.

Table 4. Enthalpy–entropy compensation parameters for the micellisation of SLS in different (water + co-solvent) mixtures.

| % v/v co-solvent | $\Delta H^*$ (kJ·mol$^{-1}$) | $T_c$ (K) |
|------------------|-----------------------------|-------|
| Water            | $-31.61$                    | 303.3 |
| Methanol         |                              |       |
| 5                | $-29.91$                    | 300.2 |
| 10               | $-29.70$                    | 300.3 |
| 15               | $-28.48$                    | 300.3 |
| 20               | $-27.49$                    | 297.5 |
| Ethanol          |                              |       |
| 5                | $-31.51$                    | 299.3 |
| 10               | $-29.60$                    | 300.3 |
| 15               | $-29.09$                    | 300.3 |
| 20               | $-25.37$                    | 300.3 |
| Propanol         |                              |       |
| 5                | $-29.20$                    | 300.3 |
| 10               | $-25.68$                    | 300.4 |
| 15               | $-25.46$                    | 300.3 |
| Ethylene Glycol  |                              |       |
| 5                | $-30.31$                    | 300.3 |
| 10               | $-30.00$                    | 300.3 |
| 15               | $-29.33$                    | 300.3 |
| 20               | $-28.81$                    | 300.4 |
4. Conclusion

The effect of methanol, ethanol, propanol and ethylene-glycol on the micellisation of SLS at different temperatures has been studied using conductometric method. It can be concluded that the micellisation process in water–alcohol solvent mixtures depends on the nature as well as on the concentration of alcohol used. The CMC values were found to increase with increase in volume fractions of methanol and ethylene-glycol in the mixed solvent system, while the values were found to decrease with an increase in the volume fraction of ethanol and propanol in the mixed solvent system with the increase in CMC values following the trend methanol > ethylene-glycol > ethanol > propanol.

The thermodynamic studies revealed that the $\Delta G_M^o$ values are generally negative in the presence of the alcohols but less negative than the value in pure water, indicating reduced spontaneity in micelle formation in the water–alcohol mixed solvent systems. The result also showed that the micellisation process is entropy-driven at low concentrations of the alcohols but enthalpy-driven at high concentrations of the alcohols in water with the micellisation process showing enthalpy–entropy compensation phenomenon.

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Table S1. Thermodynamic parameters of micellization of sodium lauroylsarcosinate in different (water + cosolvent) mixtures at 288 K.

| % v/v cosolvent | $\Delta G_M$ (kJ mol$^{-1}$) | $\Delta H_M$ (kJ mol$^{-1}$) | $\Delta S_M$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{transfer}$ (kJ mol$^{-1}$) |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Water           | –30.02          | 10.06           | 139.14          | –               |
| Methanol        |                 |                 |                 |                 |
| 5               | –28.43          | 8.83            | 129.39          | 1.59            |
| 10              | –28.26          | 8.31            | 126.99          | 1.76            |
| 15              | –27.11          | 8.84            | 124.84          | 2.91            |
| 20              | –26.25          | 8.27            | 119.87          | 3.77            |
| Ethanol         |                 |                 |                 |                 |
| 5               | –29.96          | 8.44            | 133.32          | 0.06            |
| 10              | –27.62          | 13.19           | 141.71          | 2.40            |
| 15              | –27.33          | 20.73           | 166.88          | 2.69            |
| 20              | –23.86          | 16.91           | 141.54          | 6.16            |
| Propanol        |                 |                 |                 |                 |
| 5               | –27.03          | 32.59           | 207.01          | 2.99            |
| 10              | –23.74          | 29.72           | 185.65          | 6.28            |
| 15              | –23.57          | 28.50           | 180.80          | 6.45            |
| Ethylene glycol |                 |                 |                 |                 |
| 5               | –28.89          | 6.23            | 121.95          | 1.13            |
| 10              | –28.62          | 6.16            | 120.76          | 1.40            |
| 15              | –28.28          | –1.97           | 91.35           | 1.74            |
| 20              | –27.91          | –5.95           | 76.25           | 2.11            |

Table S2. Thermodynamic parameters of micellization of sodium lauroylsarcosinate in different (water + cosolvent) mixtures at 293 K.

| % v/v cosolvent | $\Delta G_M$ (kJ mol$^{-1}$) | $\Delta H_M$ (kJ mol$^{-1}$) | $\Delta S_M$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{transfer}$ (kJ mol$^{-1}$) |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Water           | –30.64          | 6.36            | 126.30          | –               |
| Methanol        |                 |                 |                 |                 |
| 5               | –29.01          | 5.12            | 116.47          | 1.63            |
| 10              | –28.81          | 4.45            | 113.51          | 1.83            |
| 15              | –27.63          | 2.95            | 104.37          | 3.01            |
| 20              | –26.75          | –0.28           | 90.34           | 3.89            |
| Ethanol         |                 |                 |                 |                 |
| 5               | –30.63          | 4.35            | 119.39          | 0.01            |
| 10              | –28.29          | 6.58            | 119.02          | 2.35            |
| 15              | –28.02          | 9.82            | 129.16          | 2.62            |
| 20              | –24.40          | 9.39            | 115.30          | 6.24            |
| Propanol        |                 |                 |                 |                 |
| 5               | –27.85          | 20.49           | 164.97          | 2.79            |
| 10              | –24.43          | 18.84           | 147.65          | 6.21            |
| 15              | –24.30          | 16.61           | 139.64          | 6.34            |
| Ethylene glycol |                 |                 |                 |                 |
| 5               | –29.45          | 3.48            | 112.38          | 1.19            |
| 10              | –29.18          | 2.35            | 107.62          | 1.46            |
| 15              | –28.69          | –4.39           | 82.93           | 1.95            |
| 20              | –28.28          | –7.47           | 71.02           | 2.36            |
| % v/v cosolvent | $\Delta G_M$ (kJ mol$^{-1}$) | $\Delta H_M$ (kJ mol$^{-1}$) | $\Delta S_M$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{\text{transfer}}$ (kJ mol$^{-1}$) |
|-----------------|-----------------------------|-----------------------------|---------------------------------|----------------------------------|
| Water           | $-31.26$                   | $2.67$                      | $113.88$                        | $-$                              |
| Methanol        | $5$                         | $-29.58$                    | $1.40$                          | $103.95$                         | $1.68$                          |
|                 | $10$                        | $-29.36$                    | $0.58$                          | $100.48$                         | $1.90$                          |
|                 | $15$                        | $-28.25$                    | $-2.93$                         | $84.97$                          | $3.01$                          |
|                 | $20$                        | $-27.26$                    | $-8.83$                         | $61.85$                          | $4.00$                          |
| Ethanol         | $5$                         | $-31.22$                    | $0.26$                          | $105.63$                         | $0.04$                          |
|                 | $10$                        | $-28.91$                    | $-0.03$                         | $96.92$                          | $2.35$                          |
|                 | $15$                        | $-28.80$                    | $-1.08$                         | $93.03$                          | $2.46$                          |
|                 | $20$                        | $-25.07$                    | $1.87$                          | $90.39$                          | $6.19$                          |
| Propanol        | $5$                         | $-28.60$                    | $8.39$                          | $124.12$                         | $2.66$                          |
|                 | $10$                        | $-25.20$                    | $7.95$                          | $111.25$                         | $6.06$                          |
|                 | $15$                        | $-25.08$                    | $4.73$                          | $100.02$                         | $6.18$                          |
| Ethylene glycol | $5$                         | $-30.05$                    | $0.72$                          | $103.26$                         | $1.21$                          |
|                 | $10$                        | $-29.76$                    | $-1.45$                         | $95.01$                          | $1.50$                          |
|                 | $15$                        | $-29.15$                    | $-6.81$                         | $74.96$                          | $2.11$                          |
|                 | $20$                        | $-28.64$                    | $-8.99$                         | $65.95$                          | $2.62$                          |

Table S4. Thermodynamic parameters of micellization of sodium lauroylsarcosinate in different (water + cosolvent) mixtures at 303 K.

| % v/v cosolvent | $\Delta G_M$ (kJ mol$^{-1}$) | $\Delta H_M$ (kJ mol$^{-1}$) | $\Delta S_M$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{\text{transfer}}$ (kJ mol$^{-1}$) |
|-----------------|-----------------------------|-----------------------------|---------------------------------|----------------------------------|
| Water           | $-31.83$                    | $-1.02$                      | $101.67$                        | $-$                              |
| Methanol        | $5$                         | $-30.10$                    | $-2.31$                         | $91.71$                          | $1.73$                          |
|                 | $10$                        | $-29.88$                    | $-3.28$                         | $87.78$                          | $1.95$                          |
|                 | $15$                        | $-28.49$                    | $-8.82$                         | $64.93$                          | $2.91$                          |
|                 | $20$                        | $-27.35$                    | $-17.37$                        | $32.93$                          | $4.48$                          |
| Ethanol         | $5$                         | $-31.62$                    | $-3.83$                         | $91.73$                          | $0.21$                          |
|                 | $10$                        | $-29.23$                    | $-6.64$                         | $74.55$                          | $2.60$                          |
|                 | $15$                        | $-28.92$                    | $-11.98$                        | $55.90$                          | $2.91$                          |
|                 | $20$                        | $-25.38$                    | $-5.65$                         | $65.12$                          | $6.45$                          |
| Propanol        | $5$                         | $-29.34$                    | $-3.71$                         | $84.61$                          | $2.49$                          |
|                 | $10$                        | $-25.75$                    | $-2.94$                         | $75.30$                          | $6.08$                          |
|                 | $15$                        | $-25.39$                    | $-7.15$                         | $60.18$                          | $6.44$                          |
| Ethylene glycol | $5$                         | $-30.49$                    | $-2.03$                         | $93.92$                          | $1.34$                          |
|                 | $10$                        | $-30.10$                    | $-5.26$                         | $82.00$                          | $1.73$                          |
|                 | $15$                        | $-29.43$                    | $-9.23$                         | $66.67$                          | $2.40$                          |
|                 | $20$                        | $-28.93$                    | $-10.51$                        | $60.81$                          | $2.90$                          |
Table S5. Thermodynamic parameters of micellization of sodium lauroylsarcosinate in different (water + cosolvent) mixtures at 308 K.

| % v/v cosolvent | $\Delta G_M^o$ (kJ mol$^{-1}$) | $\Delta H_M^o$ (kJ mol$^{-1}$) | $\Delta S_M^o$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{transfer}$ (kJ mol$^{-1}$) |
|-----------------|-------------------------------|-------------------------------|--------------------------------------|-----------------------------------|
| Water           | −32.29                        | −4.71                         | 89.53                                | –                                 |
| Methanol        |                               |                               |                                      |                                   |
| 5               | −30.50                        | −6.03                         | 79.45                                | 1.79                              |
| 10              | −30.27                        | −7.15                         | 75.07                                | 2.02                              |
| 15              | −28.77                        | −14.70                        | 45.66                                | 3.52                              |
| 20              | −27.44                        | −25.92                        | 4.93                                 | 4.85                              |
| Ethanol         |                               |                               |                                      |                                   |
| 5               | −32.07                        | −7.91                         | 78.43                                | 0.22                              |
| 10              | −29.50                        | −13.25                        | 52.75                                | 2.79                              |
| 15              | −29.13                        | −22.88                        | 20.28                                | 3.16                              |
| 20              | −25.60                        | −13.17                        | 40.34                                | 6.69                              |
| Propanol        |                               |                               |                                      |                                   |
| 5               | −29.37                        | −15.80                        | 44.04                                | 2.92                              |
| 10              | −25.84                        | −13.82                        | 39.02                                | 6.45                              |
| 15              | −25.43                        | −19.04                        | 60.18                                | 6.86                              |
| Ethylene glycol |                               |                               |                                      |                                   |
| 5               | −30.93                        | −4.79                         | 84.88                                | 1.36                              |
| 10              | −30.49                        | −9.06                         | 69.57                                | 1.80                              |
| 15              | −29.78                        | −11.64                        | 58.90                                | 2.51                              |
| 20              | −29.21                        | −12.03                        | 55.80                                | 3.08                              |

Table S6. Thermodynamic parameters of micellization of sodium lauroylsarcosinate in different (water + cosolvent) mixtures at 313 K.

| % v/v cosolvent | $\Delta G_M^o$ (kJ mol$^{-1}$) | $\Delta H_M^o$ (kJ mol$^{-1}$) | $\Delta S_M^o$ (J mol$^{-1}$ K$^{-1}$) | $\Delta G_{transfer}$ (kJ mol$^{-1}$) |
|-----------------|-------------------------------|-------------------------------|--------------------------------------|-----------------------------------|
| Water           | −32.69                        | −8.40                         | 77.61                                | –                                 |
| Methanol        |                               |                               |                                      |                                   |
| 5               | −30.86                        | −9.74                         | 67.46                                | 1.83                              |
| 10              | −30.58                        | −11.01                        | 62.53                                | 2.11                              |
| 15              | −29.01                        | −20.59                        | 26.91                                | 3.68                              |
| 20              | −27.48                        | −34.47                        | −22.32                               | 5.21                              |
| Ethanol         |                               |                               |                                      |                                   |
| 5               | −32.48                        | −12.00                        | 65.41                                | 0.21                              |
| 10              | −29.84                        | −19.86                        | 31.88                                | 2.85                              |
| 15              | −29.26                        | −33.78                        | −14.44                               | 3.43                              |
| 20              | −25.80                        | −20.69                        | 16.33                                | 6.89                              |
| Propanol        |                               |                               |                                      |                                   |
| 5               | −29.63                        | −27.90                        | 5.52                                 | 3.06                              |
| 10              | −26.06                        | −24.71                        | 4.30                                 | 6.63                              |
| 15              | −25.64                        | −30.92                        | −16.85                               | 7.05                              |
| Ethylene glycol |                               |                               |                                      |                                   |
| 5               | −31.36                        | −7.54                         | 76.11                                | 1.33                              |
| 10              | −30.86                        | −12.87                        | 57.49                                | 1.83                              |
| 15              | −30.05                        | −14.06                        | 51.07                                | 2.66                              |
| 20              | −29.51                        | −13.55                        | 51.00                                | 3.18                              |