



Ternary diffusion in aqueous sodium salicylate + sodium dodecyl sulfate solutions

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ABSTRACT

A Taylor dispersion method has been used to measure ternary mutual diffusion coefficients (D_{11} , D_{22} , D_{12} and D_{21}) in aqueous sodium salicylate (NaSal) (component 1) + sodium dodecyl sulfate (NaDS) (component 2) solutions at 25.00 °C and concentrations up to 0.050 mol dm⁻³. In general, the data show that diffusion of NaDS drives co-current flow of NaSal, and that diffusion of NaSal drives also co-current flows of NaDS. The experimental ternary diffusion coefficients are compared with Nernst-Planck coefficients allowing a better interpretation of the electrostatic mechanism for the coupled diffusion of NaSal and SDS. From these equations, at compositions below the critical micelle concentration (cmc) of NaDS, small positive values of D_{12} and D_{21} are obtained resulting from fully dissociation of NaDS; however, at compositions above the cmc, the coupled diffusion of NaSal and NaDS becomes significant, as indicated by the experimental and predicted large positive cross-diffusion coefficients, D_{21} . There is a good agreement between our data and those predicted by the Nernst-Planck equations for NaDS concentrations below and above the cmc.

1. Introduction

Sodium salicylate (NaSal) is the deacetylated form of a well-known drug: aspirin; for that reason, the analgesic effect of NaSal is smaller; however, NaSal can be administrated to those patients showing intolerance to salicylic acid [1]. Besides that, NaSal also acts as antipyretic, non-steroidal anti-inflammatory drug [2–3] and induces apoptosis in different cancer cells, such as colon, lung, and breast cancers [4–5].

However, administration of high doses of salicylate may induce several diseases including hearing loss and neurotoxicity resulting in hyperactivity or hearing loss as a consequence of interaction between NaSal and γ -aminobutyric acid [6–8].

Although soluble in water (ca. 700 mg/mL, at 25 °C [9]), NaSal shows amphiphilic properties due to its aromatic ring, quantified by a relatively low octanol/water partition coefficient (–1.43 [10]), being classified as hydrotrope. These compounds have the ability to increase the solubility of poorly water soluble drugs [11,12]. Thus, the use of hydrotropes in chemical and pharmaceutical formulations containing

surfactants and polymers leads to a synergistic effect with a significant modification of their properties (e.g., aggregation and viscosity) [13].

The interaction between NaSal and surfactants leads to a change in micellization properties of the latter [14,15]. This can be justified by the occurrence of the screening effect due to an increase in the ionic strength -the salicylate anion decreases the electrostatic repulsion between surfactant's head-groups- favoring the micelle formation and forming mixed micelles, once the aromatic ring interacts with the hydrophobic tails of surfactants [16–17]. This also induces the occurrence of wormlike micelles. It is worth noticing that the interaction between alkyl-trimethyl ammonium surfactants and NaSal may drive the formation of different micelle phase transitions. For example, for dodecyl trimethylammonium bromide and tetramethylammonium bromide the formation of spherical and linear and branched wormlike micelles have been reported [18–20]; however, for hexadecyltrimethylammonium bromide [15,21] no spherical micelles are formed which highlights the importance of the alkyl chain length on the interaction with salicylate anion. On the other hand, only few papers on interactions between anionic

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surfactants and hydrotropes have been published so far [22,23]. Here we focus our study in the interactions between NaSal and SDS. This surfactant is the most used one in industrial applications and as a surfactant model. By using electrical conductivity measurements, Sheikh et al. [22] have found that by increasing the NaSal concentration the NaDS critical micelle concentration (cmc) decreases and the interaction between the hydrotrope and the surfactant increases, suggesting the occurrence of mixed micelles with increasing stability, enhanced by the role of the hydrophobic hydration in the micellization process; i.e., salicylate anion increases the electrostatic repulsion between DS^- head-groups, thereby disfavoring micelle formation and the formation of mixed micelles.

In this manuscript we focus our attention on the interactions between SDS and NaSal, aiming two different issues: to investigate the interdiffusion coefficient of sodium salicylate in the presence of the anionic surfactant SDS at pre- and post-micelle concentration and second to contribute for the elucidation of interaction mechanism and how they affect the transport phenomena in applied systems. For that we are using the Taylor dispersion technique (a powerful technique to measure interdiffusion coefficients in multicomponent systems).

The present work provides experimental and theoretical ternary diffusion values for NaSal and NaDS at different compositions (pre-and post-micelle concentration) by using this experimental technique and the Nernst-Planck (NP) equations [24], which predicts the multicomponent diffusion coefficients for solutions micellar electrolytes with added salt [25]. The good agreement found between these values suggests that there is no significant evidence for mixed NaSal-NaDS micelle formation, contrary to what would be expected. Instead, added NaSal lowers the cmc by the well-known common ion effect. In other words, knowing that in this system there are micelle formation reaction $50Na^+ + 60DS^- \rightleftharpoons (Na_{50}DS_{60})^{10-}$, being the critical micelle concentration (cmc) of NaDS $0.0083 \text{ mol dm}^{-3}$ [26], added Na^+ ions from added NaSal shift the equilibrium to the right, in favour of micelle formation, which in turn reduces the cmc.

Support for this phenomenon is shown in Fig. 1, which provides further evidence that neither NaSal or NaCl are solubilized by NaDS micelles. In fact, for two aqueous systems $\{NaSal (C_1) + NaCl (C_2)\}$, and $\{NaSal (C_1) + NaDS (C_2)\}$, there is a good agreement between the experimental and predicted cmc, having been considered in this discussion the absence of the NaCl-NaDS mixed micelles formation [26,27]. Moreover, the reduction in the NaDS cmc caused by added NaSal is similar to that observed in the presence of sodium chloride, suggesting the occurrence of no NaCl-NaDS mixed micelles.

2. Experimental

2.1. Materials

Table 1 describes all reagents used as received in the present work. Solutions for the diffusion measurements were prepared at room temperature (296.15 K) before each experiment in calibrated volumetric flasks using Millipore Q water (specific conductivity less than $30 \mu\text{S m}^{-1}$, at 298.15 K). The changes in the solute molarities caused by heating the solutions to 298.15 K (the temperature for the diffusion measurements) were negligible (-0.04%) compared to the uncertainties in the

Table 1
Reagents used in the experiments.

Chemical name	Source	CAS Number	Mass fraction Purity ^a
Sodium salicylate	Panreac	54-21-7	>0.99
Sodium dodecyl sulfate	Merck	151-21-3	>0.99
Water	Millipore-Q water (specific conductivity less than $30 \mu\text{S m}^{-1}$ at 298.15 K)	7732-18-5	

^a As declared by the supplier.

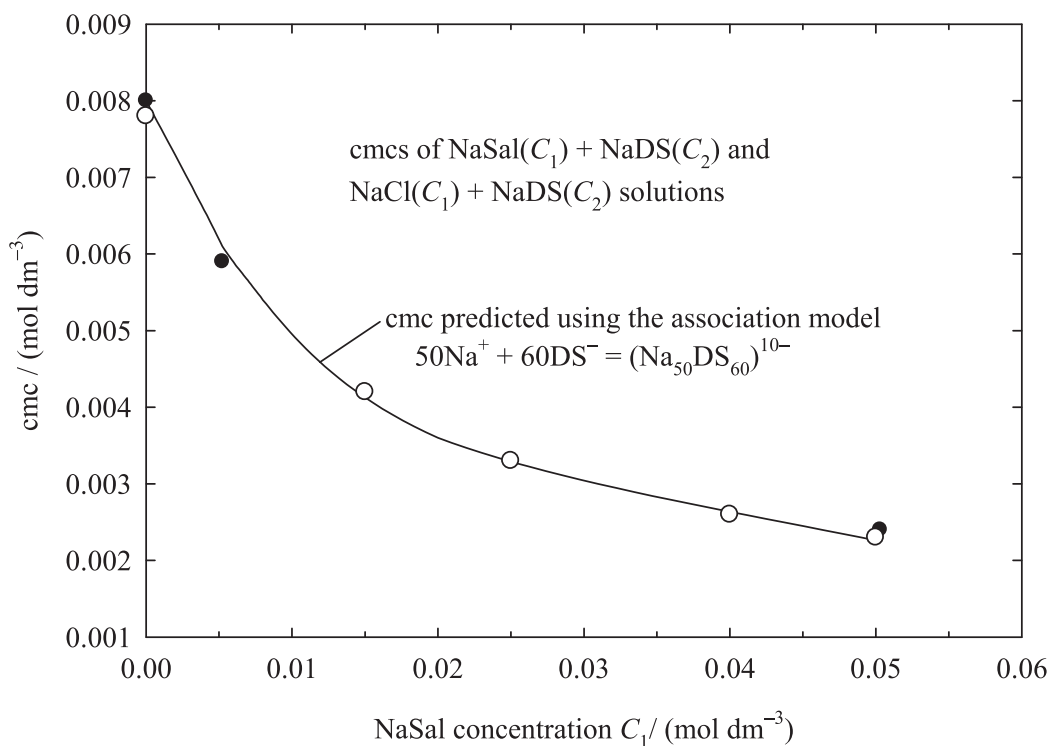


Fig. 1. Comparison of measured and predicted critical micelle concentrations (cmc) of aqueous sodium salicylate (C_1) + sodium dodecyl sulfate (C_2) solutions (filled circles, [28]), and for sodium chloride (C_1) + sodium dodecyl sulfate (C_2) solutions (open circles, [27,28]). Predicted values: solid curve; for further details check section 4.

concentrations from impurities in the NaSal and SDS samples used to prepare the solutions.

2.2. Diffusion measurements using the Taylor dispersion technique

Mutual diffusion coefficients for the mixed electrolyte solutions {NaSal(1) + NaDS(2) + water} are described by the coupled Fick's equations [24]:

$$J_1 = -D_{11}\nabla C_1 - D_{12}\nabla C_2 \quad (1)$$

$$J_2 = -D_{22}\nabla C_2 - D_{21}\nabla C_1 \quad (2)$$

where the main ternary mutual diffusion coefficients D_{11} and D_{22} represent the flux of each component (1) and (2) produced by its own concentration gradient, and the cross-diffusion coefficients D_{12} and D_{21} represent the flux of NaSal caused by the NaDS concentration gradient (∇C_2) and the flux of NaDS caused by the concentration gradient of NaSal (∇C_1), respectively.

These parameters (D_{11} , D_{12} , D_{21} and D_{22}) were measured by the Taylor dispersion technique. Once this technique is well described in the literature [29–31], we just summarize relevant points regarding the equipment and the method. At the start of each run a 0.063 cm³ sample of solution was injected into a laminar carrier solution of slightly different composition at the entrance to a Teflon capillary dispersion tube of length 3048.0 (± 0.1) cm, and internal radius 0.03220 \pm (0.00003) cm. This tube and the injection valve were kept at 298.15 (± 0.01) K in an air thermostat. The broadened distribution of the disperse samples was monitored at the tube outlet by a differential refractometer (Waters model 2410). The refractometer output voltages, $V(t)$, were measured at 5 s intervals by a digital voltmeter (Agilent 34401 A).

The dispersion profiles for these mixed electrolyte solutions (NaSal (1) + NaDS(2)) were analyzed by fitting the following equation (Eq. (3)).

$$V(t) = V_0 + V_1 t + V_{\max} \left(\frac{t_{\text{R}}}{t} \right)^{\frac{1}{2}} \left[W_1 \exp \left(-\frac{12D_1(t - t_{\text{R}})^2}{r^2 t} \right) + (1 - W_1) \exp \left(-\frac{12D_2(t - t_{\text{R}})^2}{r^2 t} \right) \right] \quad (3)$$

V_0 , V_1 and V_{\max} represent baseline voltage, baseline slope and the peak height relative to the linear baseline voltage $V_0 + V_1 t$, respectively, and t_{R} is the mean sample retention time. D_1 and D_2 , are the eigenvalues of the ternary diffusion coefficient matrix. Ternary dispersion profiles were prepared by injecting NaSal(1) + NaDS (2) solution samples of composition $\bar{C}_1 + \Delta C_1$, $\bar{C}_2 + \Delta C_2$ into carrier streams of composition \bar{C}_1, \bar{C}_2 . For flow solutions containing only one component (e.g., $C_1 \neq 0$ and $C_2 = 0$), these data were obtained by injecting solution samples of composition $\bar{C}_1 + \Delta C_1$, $\bar{C}_2 + \Delta C_2$ into carrier solutions of composition \bar{C}_1 .

Table 2

Ternary mutual diffusion coefficients, D_{11} , D_{12} , D_{21} and D_{22} , and the respective standard deviations, S_D , of aqueous NaSal (C_1) + NaDS ($C_2 < \text{cmc}$)^a solutions at 298.15 K and 101.3 kPa.

C_1^b	m_1^c	C_2^b	m_2^c	X_1^d	$D_{11} \pm S_D^e$	$D_{12} \pm S_D^e$	$D_{21} \pm S_D^e$	$D_{22} \pm S_D^e$
0.0000	0.000000	0.0040	0.00401 ₅	0.0000	0.667 \pm 0.006	0.045 \pm 0.025	0.368 \pm 0.005	0.700 \pm 0.014
0.0010	0.00100 ₁	0.0030	0.00301 ₁	0.2500	0.699 \pm 0.006	0.029 \pm 0.020	0.055 \pm 0.030	0.714 \pm 0.025
0.0020	0.00200 ₂	0.0020	0.00200 ₃	0.5000	0.781 \pm 0.023	0.076 \pm 0.020	0.165 \pm 0.033	0.739 \pm 0.025
0.0030	0.00300 ₆	0.0010	0.00100 ₃	0.7500	0.865 \pm 0.010	0.130 \pm 0.030	0.080 \pm 0.024	0.687 \pm 0.014
0.0040	0.00400 ₄	0.0000	0.000000	1.000	0.936 \pm 0.015	0.160 \pm 0.056	0.034 \pm 0.030	0.640 \pm 0.020

^a The information that these compositions are below the cmc can be inferred from the analysis of Figure 1. ^b C_1 in units of mol dm⁻³. ^cMolalities, m_i , are given per 1 kg of water (mol kg⁻¹). Molalities, m_1 and m_2 , of aqueous solutions of NaSal and NaDS, respectively, were calculated from our molarities and density measurements available in [34,35]. ^d $X_1 = C_1/(C_1 + C_2)$ represents the NaSal solute mole fraction. ^e $D_{ij} \pm S_D$ is the mean diffusion coefficient from 4 to 6 replicate measurements in units of 10⁻⁹ m² s⁻¹ and S_D is the standard deviation of that mean. Relative standard uncertainty, u_r , and standard uncertainties, u , are $u_r(C) = 0.03$; $u(T) = 0.01$ K and $u(p) = 2.03$ kPa.

In the present work, the D_{ik} values, shown in Table 2, at dilute solutions containing only one component (that is, $C_1 = 0.000$ and $C_2 = 0.004$ mol dm⁻³) were obtained by injected two samples solutions of different composition: 1) $\Delta C_1 = 0.010$ mol dm⁻³, $\Delta C_2 = 0$, and 2) $\Delta C_1 = 0$, $\Delta C_2 = 0.004$ mol dm⁻³. For solutions containing now $C_1 = 0.004$, $C_2 = 0.000$ mol dm⁻³, the compositions of two injected solutions were: 1) $\Delta C_1 = 0.010$ mol dm⁻³, $\Delta C_2 = 0$; 2) $\Delta C_1 = 0$ mol dm⁻³, $\Delta C_2 = 0.005$ mol dm⁻³. Regarding the D_{ik} values, presented in Table 3, obtained in solutions containing only $C_2 = 0.050$ mol dm⁻³, the following injection solutions were used: 1) $\Delta C_1 = 0.020$ mol dm⁻³, $\Delta C_2 = 0$; 2) $\Delta C_1 = 0$, $\Delta C_2 = 0.010$ mol dm⁻³. Details of the analysis of the dispersion profiles are given in the literature [32,33].

3. Results

Table 2 shows the ternary mutual diffusion coefficients for dilute aqueous solutions containing $(C_1 + C_2) = 0.004$ mol dm⁻³, at NaDS pre-micelle concentrations. The guarantee that these compositions are below the cmc can be inferred from the analysis of Fig. 1.

The reported mutual diffusion coefficients are the average of four to six replicate D_{ik} measurements at each composition. The main diffusion coefficients D_{11} and D_{22} were generally reproducible within $\pm 0.02 \times 10^{-9}$ m² s⁻¹. The cross-coefficients were reproducible within $\pm 0.05 \times 10^{-9}$ m² s⁻¹.

Table 3

Ternary mutual diffusion coefficients D_{11} , D_{12} , D_{21} and D_{22} , and the respective standard deviations, S_D , of aqueous NaSal (C_1) + NaDS ($C_2 = 0.0500$ mol·dm⁻³)^a solutions at 298.15 K above the cmc^b.

C_1^c	m_1^d	X_1^e	$D_{11} \pm S_D^f$	$D_{12} \pm S_D^f$	$D_{21} \pm S_D^f$	$D_{22} \pm S_D^f$
0.0000	0.00000	0.0000	0.906 \pm 0.003	0.009 \pm 0.003	0.449 \pm 0.099	0.460 \pm 0.003
0.0025	0.00250 ₂	0.0476	0.897 \pm 0.005	-0.006 \pm 0.001	0.441 \pm 0.070	0.490 \pm 0.006
0.0050	0.00500 ₂	0.0909	0.971 \pm 0.004	0.014 \pm 0.004	0.446 \pm 0.012	0.496 \pm 0.002
0.0075	0.00750 ₄	0.1304	0.993 \pm 0.010	-0.009 \pm 0.001	0.365 \pm 0.034	0.495 \pm 0.013

^a Molality of this NaDS solution, given per 1 kg of water is $m_2 = 0.05001$ mol·kg⁻¹.

^b The information that these compositions are below the cmc can be inferred from the analysis of Figure 1. ^c C_1 in units of mol dm⁻³. ^dMolalities, m_i , are given per 1 kg of water (mol kg⁻¹). Molalities (m_1) of aqueous solutions of NaSal were calculated from our molarities and density measurements available in References [34,35]. ^e $X_1 = C_1/(C_1 + C_2)$ represents the NaSal solute mole fraction. ^f $D_{ij} \pm S_D$ is the mean diffusion coefficients from 4 to 6 replicate measurements in units of 10⁻⁹ m² s⁻¹ and S_D is the standard deviation of that mean. Relative standard uncertainty, u_r , and standard uncertainties, u , are $u_r(C) = 0.03$; $u(T) = 0.01$ K and $u(p) = 2.03$ kPa.

In general, an increase of the NaSal solute mole fraction, defined as $X_1 = C_1/(C_1 + C_2)$, has an opposite effect on the main diffusion coefficients D_{11} and D_{22} ; i.e. D_{11} increases and D_{22} decreases by increasing the X_1 . Comparing the main (Table 2) and the binary ([34]) diffusion coefficients of NaSal, at the same experimental conditions, it can be concluded that there are changes in the rate of NaSal diffusion caused by the addition of NaDS. D_{11} is significantly smaller (by up to 34%) than the corresponding binary NaSal diffusion coefficients (D_1). The D_{11} values for the composition limits $X_1 = 0$ and $X_1 = 1$ correspond to the tracer diffusion coefficient of NaSal in supporting NaDS solutions and the binary mutual diffusion coefficient of aqueous NaSal, respectively. Similarly, the D_{22} values for $X_1 = 0$ and $X_1 = 1$ refer to the binary mutual diffusion coefficient of aqueous NaDS and the tracer diffusion coefficient of NaDS in NaSal aqueous solutions, respectively.

Comparing D_{22} values with the corresponding binary NaDS diffusion coefficients [25,26], we can also verify that D_{22} assume smaller values (by up to 18%). However, in the limit $X_1 \rightarrow 1$, D_{21} is zero because NaSal concentration gradients cannot drive coupled flows of NaDS in solutions that do not contain NaDS. Similarly, $D_{12} \rightarrow 0$ as $X_1 \rightarrow 0$.

Coupled diffusion in these solutions is not negligible. For example, from the values of D_{12}/D_{22} ratio, we can see that a mole of diffusing NaDS co-transport at most 0.25 mol of NaSal, whereas the values of D_{21}/D_{11} show that a mole of diffusing NaSal can cotransport up to 0.55 mol of NaDS.

Table 3 shows the ternary mutual diffusion coefficients D_{11} , D_{12} , D_{21} and D_{22} of aqueous NaSal + NaDS carrier solutions for concentrations of NaDS above the cmc (that is, $C_2 > 0.05 \text{ mol dm}^{-3}$).

Tables 4 and 5 show also data for this system but at compositions $C_1 + C_2 = 0.020 \text{ mol dm}^{-3}$ and $C_1 + C_2 = 0.050 \text{ mol dm}^{-3}$. It should be stressed that the cmc of NaDS in the presence of NaSal is indicated in Fig. 1.

Data shown in Tables 3-5 show that D_{11} and D_{22} increase by increasing the solute molar fraction, X_1 . However, it is noteworthy that D_{22} values measured above the cmc are significantly lower than those obtained for the pre-micelle region (Tables 2, 4, 5).

Looking to D_{12}/D_{22} ratio values, it can be concluded that a mole of diffusing NaDS co-transport at most 0.5 mol of NaSal. However, this coupled diffusion cannot be justified by the solubilization of NaSal in NaDS micelles. In fact, having in mind the tracer experimental D_{11} value ($D_{11} = 0.906 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$, Table 3), and assuming $D_{\text{Sal}^-}^0 = 0.918 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ [32] and $D_{\text{micelle}} = 0.10 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ [25], the fraction of Sal^- ions solubilized by the NaDS micelles, s , can be estimated by using equation (4),

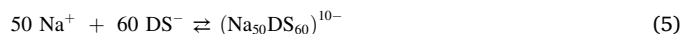
$$D_{11} = (1 - s)D_{\text{Sal}^-}^0 + sD_{\text{micelle}} \approx (1 - s)D_{\text{Sal}^-}^0 + sD_{\text{micelle}} \quad (4)$$

obtaining $s = 0.015$, that is ca. zero, indicating that the encapsulation of salicylate ions into micelles may be negligible. This fact can be also reinforced if we consider that the tracer diffusion coefficient of Sal^- ions in 0.05 mol dm^{-3} NaDS solutions (above the cmc) is $0.906 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$, does not differ significantly from the limiting $D_{\text{Sal}^-}^0 = 0.918 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ value, suggesting also that solubilization of Sal^- ions in the

slower-diffusing micelles ($D_{\text{micelle}} = 0.10 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$) is, thus, neglected. This effect is contrary to the effect of salicylate ions on the micellization of cationic surfactants as, for example, dodecyltrimethylammonium chloride, as demonstrated by Šarac et al. [36].

4. Interpretation of data using Nernst-Planck equations

For better understanding our experimental data, the ternary mutual diffusion coefficients for this system were compared with those predicted from NP equations [24], which provide a concise description of transport in terms of the fluxes of the NaSal and NaDS components. However, to interpret the results, including possible mechanisms for coupled diffusion, it is necessary to relate the fluxes of the components to the fluxes of the actual diffusing species: Sal^- , DS^- , Na^+ ions and micelles formed by the equilibrium reaction [25].



For the sake of simplicity, we are assuming the NaSD micelle aggregation number and the degree of dissociation of counter ions in micelles equal to 60 and 0.17, respectively. It should be stressed that these numbers are an approximation, once they depend on the technique used for their determination (see, for example, reference [37]).

Once the solutions investigated in the present study are dilute, in this theoretical treatment it is assumed that the ionic strength effects (e.g., screening effect [38,39]) and activity coefficient corrections can be neglected.

Considering the mass balance (Eqs. (6) and (7)), the electroneutrality condition (Eq. (8)) and mass action law equilibrium constant (Eq. (9)), the concentrations of the sodium salicylate (C_1) and sodium dodecyl sulfate (C_2) can be described in terms of concentrations of the diffusing species: Na^+ , Sal^- , DS^- , and micelles (m):

$$C_1 = c_{\text{Sal}^-} \quad (6)$$

$$C_2 = c_{\text{DS}^-} + 60c_m \quad (7)$$

$$0 = c_{\text{Na}^+} - c_{\text{Sal}^-} - c_{\text{DS}^-} - 10c_m \quad (8)$$

$$K = \frac{C_m}{C_{\text{Na}}^{50} C_{\text{DS}}^{60}} \quad (9)$$

The equilibrium constant K is evaluated by using the accurate approximation $K = c_m^{-110}$ [25] and the critical micelle concentration $\text{cmc} = 0.0083 \text{ mol dm}^{-3}$ for binary aqueous sodium dodecyl sulfate solutions.

According to the NP equations [24], the flux of each solute species s ($s = \text{Sal}^-$, Na^+ , DS^-) is the sum of two contributions: the pure diffusional flux, $j_{s(D)}$, driven by the species concentration gradient ∇c_s , and the flux $j_{s(E)}$ of species s caused by the diffusion-induced electric field (valid for charged species).

$$j_i = j_{s(D)} + j_{s(E)} \quad (10)$$

Table 4

Ternary mutual diffusion coefficients, D_{11} , D_{12} , D_{21} and D_{22} , and the respective standard deviations, S_D , of aqueous NaSal (C_1) + NaDS (C_2) solutions at 298.15 K and 101.3 kPa. $C_1 + C_2 = 0.020 \text{ mol dm}^{-3}$.

C_1^a	m_1^b	C_2^a	m_2^b	X_1	$D_{11} \pm S_D^d$	$D_{12} \pm S_D^d$	$D_{21} \pm S_D^d$	$D_{22} \pm S_D^d$
Below the cmc^e								
0.018	0.0180 ₉	0.002	0.00200 ₇	0.9000	1.160 ± 0.023	0.340 ± 0.015	-0.035 ± 0.009	0.698 ± 0.002
Above the cmc^e								
0.012	0.0120 ₇	0.008	0.00803 ₉	0.6000	1.150 ± 0.022	0.140 ± 0.0010	-0.045 ± 0.010	0.275 ± 0.001

^a C_1 in units of mol dm^{-3} . ^b Molalities, m_i , are given per 1 kg of water (mol kg^{-1}). Molalities m_1 and m_2 of aqueous solutions of NaSal and NaDS, respectively, were calculated from our molarities and density measurements available in References [34,35]. ^c $X_1 = C_1/(C_1 + C_2)$ represents the NaSal solute mole fraction. ^d $D_{ij} \pm S_D$ is the mean diffusion coefficients from 4 to 6 replicate measurements in units of $10^{-9} \text{ m}^2 \text{ s}^{-1}$ and S_D is the standard deviation of that mean. Relative standard uncertainty, u_r , and standard uncertainties, u , are $u_r(C) = 0.03$; $u(T) = 0.01 \text{ K}$ and $u(p) = 2.03 \text{ kPa}$. ^e The information that these compositions are below the cmc can be inferred from the analysis of Figure 1.

Table 5

Ternary mutual diffusion coefficients' D_{11} , D_{12} , D_{21} and D_{22} , and the respective standard deviations, S_D , of aqueous NaSal (C_1) + NaDS (C_2) at 298.15 K and 101.3 kPa. $C_1 + C_2 = 0.050 \text{ mol}\cdot\text{dm}^{-3}$.

C_1^a	m_1^b	C_2^a	m_2^b	x_1^c	$D_{11} \pm S_D^d$	$D_{12} \pm S_D^d$	$D_{21} \pm S_D^d$	$D_{22} \pm S_D^d$
0.049	0.0497 ₅	0.001	0.00100 ₃	0.9800	1.267 ± 0.013	0.399 ± 0.016	-0.030 ± 0.009	0.840 ± 0.003
0.040	0.0405 ₂	0.010	0.0100 ₅	0.8000	1.078 ± 0.022	0.020 ± 0.010	0.020 ± 0.010	0.297 ± 0.001

^a C_1 and C_2 in units of mol dm^{-3} . ^b Molalities, m_i , are given per 1 kg of water (mol kg^{-1}). Molalities m_1 and m_2 of aqueous solutions of NaSal and NaDS, respectively, were calculated from our molarities and density measurements available in References [34,35]. ^c $x_1 = C_1/(C_1 + C_2)$ represents the NaSal solute mole fraction. ^d Mean diffusion coefficient from 4 to 6 replicate measurements in units of $10^{-9} \text{ m}^2 \text{ s}^{-1}$. Relative standard uncertainty, u_r , and standard uncertainties, u , are $u_r(C) = 0.03$; $u(T) = 0.01 \text{ K}$ and $u(p) = 2.03 \text{ kPa}$.

where,

$$j_{s(D)} = -D_i \nabla C_i \quad (11)$$

$$j_{s(E)} = \left(\frac{F}{RT} \right) D_i C_i z_i E \quad (12)$$

D_i , C_i and z_i represent the concentration, the diffusion coefficient and the charge number of species s , and, E , F , R , T are the electric field, Faraday constant, gas constant and the temperature, respectively.

Mutual diffusion in dilute aqueous NaSal + NaDS solutions produces fluxes of three different solute species (Na^+ , Sal^- , DS^- monomer). Having in mind the electroneutrality condition,

$$\sum_s z_s j_s = 0 \quad (13)$$

we can consider that there are two independent diffusion fluxes. In turn, each flux of the solute component (NaSal and NaDS), J_i , used in the Fick equations can always be expressed as the sum of the fluxes of the i -containing solute species, j_s ,

Comparing the Fick equations (Eqs. (1) and (2)) for the flows of components NaSal and NaDS with the NP equations (Eqs. 10–12) for the flows of the species, and after some rearrangement, it is shown that each Fick diffusion coefficient can be the sum of two contributions (Eq. (14)): pure diffusion (Eq. (15)) and ionic transport caused by the electric field induced by the diffusion (Eq. (16)).

$$D_{ik} = D_{ik(D)} + D_{ik(E)} \quad (14)$$

$$D_{ik(D)} = \sum_s \nu_{is} D_s \left(\frac{\partial C_s}{\partial C_k} \right)_{C_{i \neq k}} \quad (15)$$

$$D_{ik(E)} = - \sum_p \sum_s \nu_{is} \left(\frac{t_s}{z_s} \right) z_p D_p \left(\frac{\partial C_s}{\partial C_k} \right)_{C_{i \neq k}} \quad (16)$$

where transference number t_s gives the fraction of the total current carried by each solute species (i.e. Sal^- , Na^+ and DS^-) (Eq. (17)) [34].

$$t_s = \frac{z_s j_{s(E)}}{\sum_p z_p j_{p(E)}} \quad (17)$$

For compositions below the cmc (no micelle formation or other ion association), D_{ik} coefficients predicted by the NP equations (Eqs. 14–17) simplify to the much simpler limiting Nernst equations (Eqs. 18–21).

$$D_{11}^0 = D_{\text{Sal}}^0 + t_{\text{Sal}} (D_{\text{Na}}^0 - D_{\text{Sal}}^0) \quad (18)$$

$$D_{12}^0 = t_{\text{Sal}} (D_{\text{Na}}^0 - D_{\text{DS}}^0) \quad (19)$$

$$D_{21}^0 = t_{\text{DS}} (D_{\text{Na}}^0 - D_{\text{Sal}}^0) \quad (20)$$

$$D_{22}^0 = D_{\text{DS}}^0 + t_{\text{DS}} (D_{\text{Na}}^0 - D_{\text{DS}}^0) \quad (21)$$

D_{Sal}^0 , D_{Na}^0 , and D_{DS}^0 are the limiting diffusion coefficients of the Sal^- , Na^+ , and DS^- ions, respectively, and t_{Sal} and t_{DS} represent the fraction of the total current carried by the Sal^- and DS^- ions, respectively. The latter parameters can be estimated by using the following equations,

$$t_{\text{Sal}} = \frac{C_1 D_{\text{Sal}}}{c_1 D_{\text{Sal}} + c_2 D_{\text{DS}} + (c_1 + c_2) D_{\text{Na}}} \quad (22)$$

and,

$$t_{\text{DS}} = \frac{C_1 D_{\text{DS}}}{c_1 D_{\text{Sal}} + c_2 D_{\text{DS}} + (c_1 + c_2) D_{\text{Na}}} \quad (23)$$

where D_{Sal} , D_{Na} and D_{DS} are the limiting diffusion coefficients of the Sal^- , Na^+ , and DS^- ions, respectively.

The diffusion coefficients of these ionic species, D_s^0 , used in the Nernst equations (Eqs. 18–23) were computed by using Eq. (24) [40],

$$D_s^0 = \frac{RT \lambda_s^0}{z_s^2 F^2} \quad (24)$$

and considering the values of the respective limiting ionic conductivities, λ_s^0 , indicated in the references [25,40,41] (Table 6).

From analysis of equations (18) and (23), it can be seen that each main coefficient is defined by the sum of a pure diffusion contribution and an electrostatic contribution from the drift of ions in the electric generated by mutual diffusion.

In contrast, cross-diffusion coefficients D_{12} and D_{21} , are purely electrostatic.

Cross-coefficient D_{21}^0 for the coupled diffusion of NaSal driven by NaSal concentration gradients is proportional to the difference $D_{\text{Na}}^0 - D_{\text{Sal}}^0$. Thus, the electric field produced by NaSal concentration gradient leads to a slowing of the Na^+ ions and, concomitantly, a co-current coupled flow of DS^- ions ($D_{\text{Sal}}^0 = 0.918 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$) is generated. This is supported by the positive D_{21}^0 values. The same is observed for the analysis of D_{12}^0 values.

In Fig. 2, the ternary diffusion coefficients (D_{ik}^0) measured (Table 2) and predicted by using Nernst equations (Eqs. 18–23) [24] for solutions at $C_1 + C_2 = 0.004 \text{ mol dm}^{-3}$ are plotted against the mole fraction of NaSal.

As shown in Fig. 2, in general, a good qualitative agreement is obtained for the predicted Nernst D_{ik}^0 compared to the experimental ones. The small deviations can be justified by the absence of any electrophoretic term in Nernst equations or by the nonideal solution behaviour. In Supplementary material, Figs. S1 and S2 allow us to analyse the behaviour of two contributions ($D_{1k(D)}^0$ and $D_{1k(E)}^0$) for each D_{ik}^0 for the same mole fraction range. That, it can be seen that the pure-diffusion contributions $D_{11(D)}^0$ and $D_{22(D)}^0$ for the main diffusion are dominant. Concerning cross-diffusion coefficient values, D_{12}^0 and D_{21}^0 , the diffusion-

Table 6

Diffusion coefficients (D_s^0) and conductivities (λ_s^0) for ionic species at infinitesimal concentration and at 298.15 K.

Species	$\lambda_s^0 / (10^{-4} \text{ S m}^2 \text{ mol}^{-1})$	$D_s^0 / (10^{-9} \text{ m}^2 \text{ s}^{-1})^d$
Na^+	50.1 ^a	1.330 ^a
Sal^-	34.5 ^b	0.918 ^b
DS^-	22.9 ^c	0.609 ^c

^a [40]. ^b [41]. ^c [25]. ^d Values estimated from Eq. (24) and using the limiting ionic conductivities here indicated.

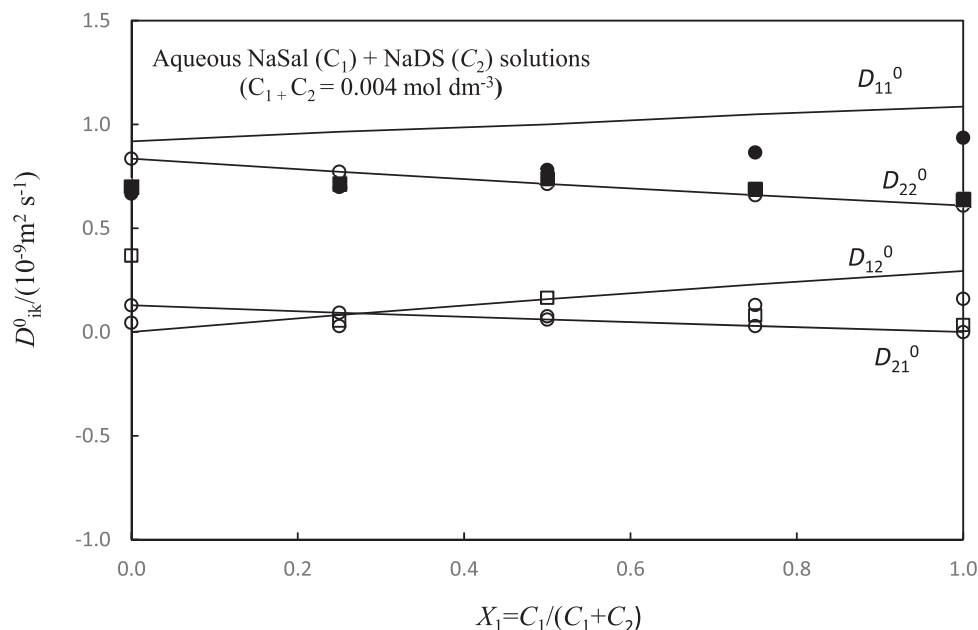


Fig. 2. Ternary diffusion coefficients D_{ik} for NaSal (C_1) + NaDS (C_2) solutions measured below the cmc, experimental values D_{11} , filled circles; D_{12} , hollow circles; D_{21} , hollow squares; D_{22} , filled squares; the solid curves represent the calculated values obtained by the Nernst model according to Eqs.18–23 [24].

induced electric fields, quantified by the positive values of $D_{12(E)}$ and $D_{21(E)}$, respectively, explain the behaviour of these transport coefficients.

The validation of this model (Eqs.14–16) was also made by the experimental determination of ternary diffusion coefficients for $C_1 + C_2 = 0.020$ and $0.050 \text{ mol dm}^{-3}$, at pre- and post-micelle concentrations (Table 7).

Table 7

Predicted ternary mutual diffusion coefficients^a, D_{ik} , and the respective pure-diffusion $D_{ij(D)}$, and electric contributions $D_{ij(E)}$ for aqueous NaSal (C_1) + NaDS (C_2) solutions.

C_1^b	C_2^b	X_1^c	D_{11}	D_{12}	D_{21}	D_{22}
($C_1 + C_2 = 0.020 \text{ mol} \cdot \text{dm}^{-3}$)						
Below the cmc^d						
0.018	0.002	0.90	1.071	0.269	0.013	0.629
			$D_{11(D)} = 0.918$	$D_{12(D)} = 0$	$D_{21(D)} = 0$	$D_{22(D)} = 0.609$
			$D_{11(E)} = 0.153$	$D_{12(E)} = 0.269$	$D_{21(E)} = 0.013$	$D_{22(E)} = 0.020$
Above the cmc^d						
0.012	0.008	0.60	1.019	0.061	-0.032	0.129
			$D_{11(D)} = 0.918$	$D_{12(D)} = 0$	$D_{21(D)} = -0.092$	$D_{22(D)} = 0.093$
			$D_{11(E)} = 0.101$	$D_{12(E)} = 0.061$	$D_{21(E)} = 0.060$	$D_{22(E)} = 0.036$
($C_1 + C_2 = 0.050 \text{ mol} \cdot \text{dm}^{-3}$)						
Below the cmc^d						
0.018	0.002	0.90	1.083	0.289	0.002	0.613
			$D_{11(D)} = 0.918$	$D_{12(D)} = 0$	$D_{21(D)} = 0$	$D_{22(D)} = 0.609$
			$D_{11(E)} = 0.165$	$D_{12(E)} = 0.289$	$D_{21(E)} = 0.002$	$D_{22(E)} = 0.004$
Above the cmc^d						
0.012	0.008	0.60	1.069	0.078	0.022	0.119
			$D_{11(D)} = 0.918$	$D_{12(D)} = 0$	$D_{21(D)} = -0.016$	$D_{22(D)} = 0.099$
			$D_{11(E)} = 0.151$	$D_{12(E)} = 0.078$	$D_{21(E)} = 0.038$	$D_{22(E)} = 0.020$

^a D_{11} , D_{12} , D_{21} and D_{22} coefficients and the respective contributions ($D_{ik(D)}$ and $D_{ik(E)}$) in units of $10^{-9} \text{ m}^2 \text{ s}^{-1}$. ^b C_1 and C_2 in units of mol dm^{-3} . ^c $X_1 = C_1 / (C_1 + C_2)$ represents the NaSal solute mole fraction. ^d The information that these compositions are below or above cmc can be inferred from the analysis of Fig. 1.

Table 7 also shows the pure-diffusion and electrostatic contributions for these D_{ik} coefficients. The values for D_{12} and D_{21} clearly indicates that pure-diffusion contributions to the coupled diffusion are negligible; however, considering the electrostatic contributions, it can be concluded that they are higher for NaDS at concentrations below the cmc. Because Na^+ ions are more mobile than DS^- ions (Table 5), the electric field along the NaDS gradients speeds up the diffusing Sal^- and, consequently, $D_{12} > 0$. Regarding the main D_{ii} coefficients, the NP analysis suggests that pure-diffusion contributions for these parameters are the most important.

The dependency of predicted D_{ik} from the referred equations (Eqs.14–16) as a function of mole fraction, for $C_1 + C_2 = 0.020 \text{ mol dm}^{-3}$, is plotted as solid lines in Fig. 3. Focusing on the main diffusion coefficient for sodium dodecyl sulfate (D_{22}), it can be seen that at the low mole fractions X_1 , D_{22} are significantly lower than those obtained for the pre-micelle region (Fig. 3), indicating the presence of micelles. A possible explanation is that as the NaDS concentration is raised above the cmc, the micelles start to form, and consequently, there is a very sharp drop in D_{22} . Because the composition is above the cmc, part of the NaDS component diffuses as the slower-moving micelles, so D_{22} is much lower than the D_{22} values below the cmc you've measured. At high mole fractions ($X_1 = 0.90$), because NaDS diffuses as Na^+ and DS^- ions (no micelles are present), a sharp D_{22} increase is noticed.

Despite the approximations assumed in these theoretical calculations, the good agreement between predicted D_{ik} coefficients and the measured D_{ik} coefficients shows the lowering of the NaDS cmc is accounted for, almost quantitatively, the well-known common-ion effect.

5. Conclusions

The effects of added sodium dodecyl sulfate on the diffusion of aqueous sodium salicylate at 298.15 K have been investigated by measuring ternary mutual diffusion coefficients for aqueous solutions of NaSal + NaDS. The measured D_{ik} coefficients have been compared with D_{ik}^0 coefficients predicted by the Nernst-Planck equations. Based on the ionic mobilities and diffusion coefficients, these equations predict small positive values of D_{12} and D_{21} below the critical micelle concentration (cmc), where ionic dissociation is complete and NaSal + NaDS solutions

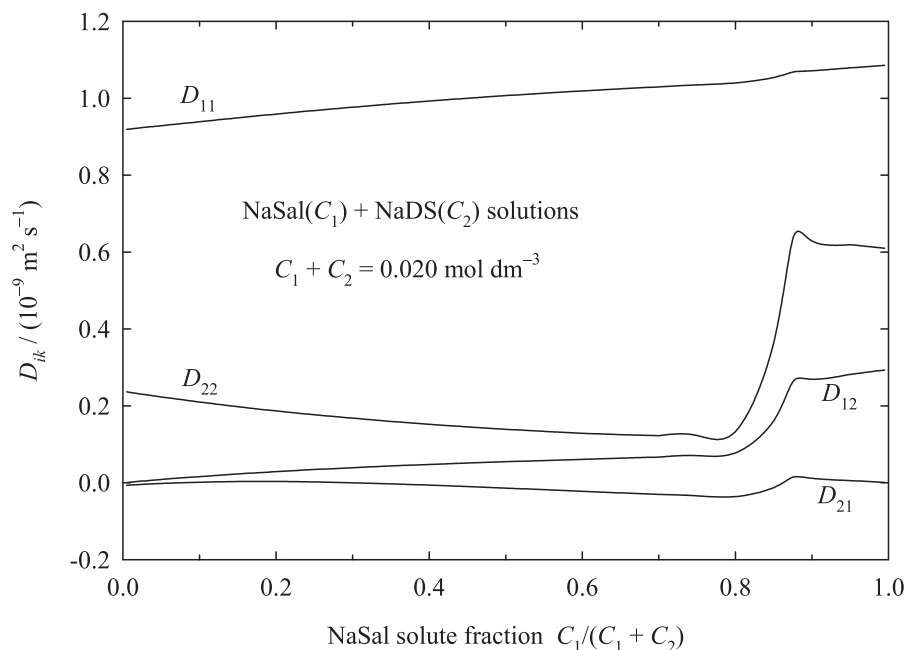


Fig. 3. The dependence of ternary D_{ik} coefficients for NaSal (C_1) + NaDS (C_2) solutions containing $0.020 \text{ mol dm}^{-3}$ total solute as calculated by using NP equations (Eqs (14) to (18)).

should behave as strong ternary mixed electrolyte.

Above the cmc, which the presence of micelles only of type $(\text{Na}_{50}\text{DS}_{60})^{10-}$ is assumed, the good agreement between our data and predicted ones suggests that the solubilization of Sal^- ions by the NaDS micelles is negligible, and so the diffusion of mixed-micelle species such as $(\text{Na}_{50}\text{Sal}_1\text{DS}_{59})^{10-}$ can be ignored without serious error.

In addition, it is also possible to conclude that the common ion effect can be considered the main reason for obtaining significant coupled fluxes of NaSal, resulting from NaDS concentration gradients in solution.

CRedit authorship contribution statement

M. Melia Rodrigo: Conceptualization, Investigation, Methodology, Software, Data curation, Writing – review & editing, Resources. **Artur J. M. Valente:** Conceptualization, Investigation, Methodology, Data curation, Writing – review & editing, Resources, Funding acquisition. **Miguel A. Estes:** Conceptualization, Investigation, Methodology, Software, Writing – original draft, Writing – review & editing, Supervision, Resources, Funding acquisition. **Ana M.T.D.P.V. Cabral:** Conceptualization, Investigation, Methodology, Software, Data curation, Writing – review & editing, Resources. **Ana C.F. Ribeiro:** Conceptualization, Investigation, Methodology, Software, Data curation, Writing – original draft, Writing – review & editing, Supervision, Resources, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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