

# Surface Tension of Dialkyl Carbonates + (Alkanes or 1,4-Dimethylbenzene) and 1,4-Dimethylbenzene + Alkanes Binary Mixtures at $T = 308.15$ K

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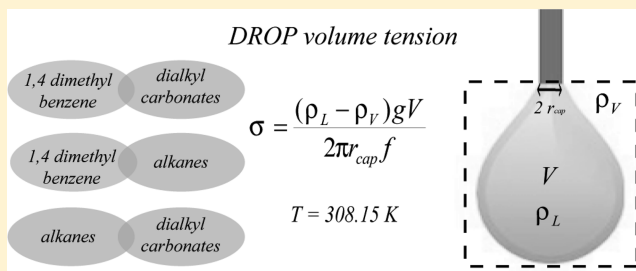
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**ABSTRACT:** New experimental surface tension data are reported for binary mixtures of dimethyl carbonate and diethyl carbonate with octane, decane, and 1,4-dimethylbenzene and 1,4-dimethylbenzene with octane and decane at 308.15 K and atmospheric pressure. These experimental results are compared with those reported in the literature at different temperature when they are available. The experimental results show that surface tension of these compounds depends systematically on temperature. The surface tension deviation calculated was fitted by the Redlich–Kister polynomial equation. The adjustable parameters obtained for each system are reported with the standard deviations between experimental and calculated values.



## INTRODUCTION

Dialkyl carbonates are nonirritating and nontoxic chemicals used in the chemical industry in applications like the synthesis of pharmaceuticals and agrochemicals, extractive processes, paint solvents, battery technology, and so forth.<sup>1–3</sup> In particular, for their excellent combustion characteristics, organic carbonates such as dimethyl (DMC) and diethyl carbonates (DEC) have been considered for their addition to fuel to replace traditional oxygenated gasoline additives such as methyl tertiary butyl ether (MTBE).<sup>4,5</sup> Also, they have been proposed as lubricants to replace chlorofluorocarbons (CFCs) with new hydrochlorofluorocarbon refrigerants (HFCs).<sup>2,6</sup> For this reason, thermophysical data of mixtures containing these alkyl carbonates with main constituents of gasoline and lubricants are of particular interest. In this sense, surface tension has an important role in several industrial process, especially influencing those that involve mass transfer phenomena, for example, distillation, absorption, chemical reaction in fluid surfaces, membrane operation, and particularly in lubrication processes.<sup>7</sup>

Thus, this work is a continuation of the studies on the thermophysical properties of binary mixtures containing dialkyl carbonates, alkanes, and alkylbenzene.<sup>8–12</sup> In particular, in this manuscript, new experimental data of the surface tension for binary mixtures of dimethyl carbonate and diethyl carbonate with octane, decane, and 1,4-dimethylbenzene and 1,4-dimethylbenzene with octane and decane at 308.15 K and

atmospheric pressure are presented. From these data, the surface tension deviation was calculated, and the results were fitted with the Redlich–Kister polynomial equation.<sup>13</sup> The influence of the chain length of dialkyl carbonate and alkanes as well as the temperature effects on this property were also analyzed.

## EXPERIMENTAL SECTION

The components employed in this work were: decane ( $\geq 0.99$  mole fraction) supplied by Sigma Aldrich, and octane ( $\geq 0.995$  mole fraction), dimethyl carbonate ( $\geq 0.995$  mole fraction), diethyl carbonate ( $\geq 0.995$  mole fraction), and 1,4-dimethylbenzene ( $\geq 0.99$  mole fraction) supplied by Fluka. Until their use, the chemicals were stored under sun and humidity protection conditions. These products were not subjected to further purification processes. For the sample preparation, liquids were weighed using a Mettler AE-240 Delta Range balance with a precision of  $\pm 5 \cdot 10^{-5}$  g. With the purpose to avoid losses by evaporation, the compounds were weighed in increasing order of volatility. The uncertainty in the mole fractions of the prepared mixtures was estimated as  $\pm 10^{-4}$ .

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Table 1. Experimental Information Available of Surface Tension of Pure Components and Their Binary Mixtures<sup>a</sup>

pure components	T/K	$\sigma/\text{mN}\cdot\text{m}^{-1}$	
		this work	ref.
dimethyl carbonate	298.15		28.63; <sup>17</sup> 28.58; <sup>17</sup> 28.08 <sup>10</sup>
	308.15	26.55	
	313.15		26.62; <sup>17</sup> 26.57 <sup>17</sup>
	288.15 to 333.15		29.93 to 23.88 <sup>18</sup>
	282.11 to 371.32		30.7 to 17.8 <sup>1</sup>
diethyl carbonate	298.15		25.92; <sup>17</sup> 25.87; <sup>17</sup> 25.43 <sup>9</sup>
	308.15	24.02	24.00 <sup>19</sup>
	313.15		24.26; <sup>17</sup> 24.22 <sup>17</sup>
	288.15 to 363.15		26.97 to 18.72 <sup>18</sup>
	273.18 to 373.17		29.2 to 18.0 <sup>19</sup>
1,4-dimethylbenzene	298.15		27.43; <sup>9,10</sup> 27.70; <sup>20</sup> 27.76; <sup>21–23</sup> 27.80; <sup>24</sup> 27.89; <sup>25</sup> 28.00; <sup>21</sup> 28.02; <sup>18</sup> 28.08; <sup>26</sup> 28.11 <sup>27</sup>
	303.15		27.59; <sup>27</sup> 27.47 <sup>26</sup>
	308.15	26.29	
	296.05 (19.3 kPa)		28.27 <sup>28</sup>
	307.90 (38.7 kPa)		26.89 <sup>28</sup>
octane	333.75 (46.7 kPa)		24.21 <sup>28</sup>
	298.15		20.99; <sup>10</sup> 21.14; <sup>18</sup> 21.17; <sup>29</sup> 21.18; <sup>21</sup> 21.19 <sup>30</sup>
	308.15	19.9	
	293.15 to 513.15		21.82 to 3.10 <sup>31</sup>
	298.15 to 393.15		21.26 to 12.60 <sup>32</sup>
decane	273.15 to 393.15		23.52 to 14.01 <sup>33</sup>
	273.15 to 333.15		23.75 to 17.85 <sup>34</sup>
	273.15 (10.7 kPa)		23.88 <sup>28</sup>
	307.62 (13.9 kPa)		20.44 <sup>28</sup>
	333.15 (75.5 kPa)		17.92 <sup>28</sup>
	293.15		23.83; <sup>35</sup> 24.12; <sup>18</sup> 24.47 <sup>36</sup>
	298.15		23.39; <sup>37</sup> 23.26; <sup>30</sup> 23.37; <sup>38,21</sup> 23.10 <sup>9</sup>
	303.15		22.91; <sup>35</sup> 23.16; <sup>18</sup> 23.35 <sup>36</sup>
	308.15	22.10	22.43 <sup>37</sup>
	313.15		21.91; <sup>35</sup> 22.22; <sup>18</sup> 22.45 <sup>36</sup>
323.15		21.07; <sup>35</sup> 21.17; <sup>18</sup> 21.55 <sup>36</sup>	
333.15		20.15; <sup>35</sup> 20.29; <sup>18</sup> 20.60 <sup>36</sup>	
343.15		19.23; <sup>35</sup> 19.4; <sup>18</sup> 19.70 <sup>36</sup>	
273.15 to 393.15		25.67 to 16.47 <sup>33</sup>	
273.15 to 333.15		25.73 to 20.22 <sup>34</sup>	
binary mixtures			T/K ref.
diethyl carbonate + decane			298.15 9
diethyl carbonate + 1,4-dimethylbenzene			298.15 9
1,4-dimethylbenzene + decane			298.15 9

<sup>a</sup>Pressure differences from atmospheric pressure are indicated in parentheses.

An automatic tensiometer Lauda TVT2 was used for the determination of surface tension of pure compounds and binary mixtures which is based on the principle of the pending drop volume. This method consists of an exact determination of the volume of the drop, which is continuously formed at the end of a capillary, by a precise dosing system. The drop volume is measured during the time interval of droplet formation. This time interval is determined by a light barrier, and it goes from the detaching of a drop from the capillary end to the next one. The measuring device is the same than the previous model TVT1 described by Miller et al.<sup>14</sup> which basically differs only in the control unit.

The surface tension measurement by the drop volume method can be applied to liquid–gas and liquid–liquid interfaces without modifications. This fact represents an advantage regarding the ring detachment or Wilhelmy plate methods, where disturbing and wetting effects are observed. Furthermore, this method requires small sample quantities, and the temperature control of the measurement can be set in a wide temperature range.<sup>15</sup>

The surface tension is determined by the following equation:

$$\sigma = \frac{\Delta\rho gV}{2\pi r_{\text{cap}} f} \quad (1)$$

where  $\Delta\rho$  is the density difference between the liquid and vapor phase,  $g$  is the gravitational acceleration,  $V$  is the drop volume,  $2r_{\text{cap}}$  is the outer diameter of the capillary, and  $f$  is a shape correction factor. The TVT2 software determines the correction factor  $f$  using a polynomial function proposed by Wilkinson.<sup>16</sup>

The total accuracy of the surface tension determination is better than  $0.1 \text{ mN}\cdot\text{m}^{-1}$  and depends on the precision of the measurements of the capillary diameter, drop volume, and the density difference between liquid and vapor phases.

## RESULTS AND DISCUSSION

Experimental results of surface tension ( $\sigma$ ) were measured for dimethyl carbonate, diethyl carbonate, octane, decane, and

**Table 2. Experimental Surface Tension ( $\sigma$ ) and Surface Tension Deviation ( $\Delta\sigma$ ) of Dimethyl Carbonate with Octane and Decane, Diethyl Carbonate with Octane and Decane, and 1,4-Dimethylbenzene with Octane and Decane at 308.15 K and Atmospheric Pressure<sup>a</sup>**

$x_1$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma/\text{mN}\cdot\text{m}^{-1}$	$x_1$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma/\text{mN}\cdot\text{m}^{-1}$	$x_1$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma/\text{mN}\cdot\text{m}^{-1}$	$x_1$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\Delta\sigma/\text{mN}\cdot\text{m}^{-1}$
Dimethyl Carbonate (1) + Octane (2)			Dimethyl Carbonate (1) + Decane (2)			Dimethyl Carbonate (1) + 1,4-Dimethylbenzene (2)			Diethyl Carbonate (1) + 1,4-Dimethylbenzene (2)		
0.0503	19.80	-0.443	0.0512	21.95	-0.381	0.2988	25.84	-0.525	0.2985	25.42	-0.194
0.0976	19.78	-0.770	0.0900	21.92	-0.582	0.4013	25.76	-0.638	0.3971	25.18	-0.211
0.1960	19.85	-1.354	0.2006	21.90	-1.092	0.5016	25.70	-0.715	0.4956	24.94	-0.224
0.2941	19.93	-1.929	0.2853	21.92	-1.446	0.6032	25.67	-0.772	0.5433	24.83	-0.226
0.3969	20.09	-2.454	0.3971	21.97	-1.897	0.7029	25.67	-0.803	0.6007	24.70	-0.225
0.4950	20.25	-2.942	0.4949	22.11	-2.190	0.7964	25.74	-0.751	0.6457	24.60	-0.227
0.5455	20.39	-3.139	0.5987	22.01	-2.757	0.8945	25.98	-0.541	0.6995	24.49	-0.215
0.5971	20.54	-3.332	0.6988	22.18	-3.034	0.9442	26.13	-0.401	0.7986	24.30	-0.177
0.6977	20.80	-3.735	0.7971	22.54	-3.105				0.8990	24.12	-0.133
0.7977	21.47	-3.729	0.8983	23.81	-2.291				0.9497	24.06	-0.074
0.8983	23.04	-2.833	0.9505	24.82	-1.515						
0.9492	24.53	-1.681									
Diethyl Carbonate (1) + Octane (2)			Diethyl Carbonate (1) + Decane (2)			1,4-Dimethylbenzene (1) + Octane (2)			1,4-Dimethylbenzene (1) + Decane (2)		
0.0490	19.83	-0.274	0.0512	21.90	-0.300	0.0488	20.02	-0.191	0.1047	22.18	-0.368
0.1008	19.89	-0.429	0.0983	21.84	-0.453	0.1090	20.25	-0.355	0.2152	22.39	-0.621
0.2000	19.91	-0.821	0.1997	21.75	-0.731	0.1953	20.52	-0.632	0.2977	22.57	-0.781
0.2969	20.05	-1.075	0.2968	21.78	-0.888	0.2973	20.96	-0.839	0.3945	22.79	-0.971
0.3972	20.26	-1.279	0.4060	21.84	-1.041	0.3959	21.41	-1.021	0.4993	23.11	-1.091
0.4971	20.50	-1.447	0.4990	21.88	-1.180	0.4978	21.91	-1.170	0.5490	23.28	-1.128
0.5495	20.63	-1.537	0.5621	21.96	-1.225	0.5503	22.20	-1.222	0.5997	23.46	-1.165
0.5991	20.79	-1.580	0.5994	21.99	-1.261	0.5935	22.46	-1.234	0.7004	23.88	-1.162
0.6950	21.16	-1.610	0.6966	22.19	-1.254	0.6966	23.11	-1.246	0.8018	24.47	-0.994
0.7938	21.70	-1.474	0.7951	22.49	-1.141	0.8004	23.93	-1.084	0.9004	25.20	-0.679
0.8978	22.63	-0.969	0.9018	23.10	-0.734	0.9017	24.92	-0.739	0.9501	25.67	-0.420
0.9504	23.33	-0.489	0.9511	23.50	-0.435	0.9504	25.50	-0.475			
Dimethyl Carbonate (1) + 1,4-Dimethylbenzene (2)			Diethyl Carbonate (1) + 1,4-Dimethylbenzene (2)								
0.0503	26.19	-0.115	0.0995	25.98	-0.088						
0.1937	25.97	-0.369	0.2009	25.68	-0.158						

<sup>a</sup>The uncertainty of the variables are:  $u(T) = 5.10^{-2}$  K;  $u(x_1) = \pm 10^{-4}$ ;  $u(\sigma) = 0.1$  mN·m<sup>-1</sup>; and the combined expanded uncertainty with level of confidence 0.95 ( $k = 2$ ) for the superficial tension is:  $U_c(\sigma) = 0.2$  mN·m<sup>-1</sup>.

**Table 3. Redlich–Kister Coefficients and Standard Deviations ( $S$ )\* of eq 3 for  $\Delta\sigma$  for the Investigated Systems at 308.15 K**

$A_0$	$A_1$	$A_2$	$A_3$	$S$
-11.6586	-9.5040	-12.4234	-6.8395	0.031
-9.0475	-8.4562	-11.5584	-5.1802	0.046
-5.8643	-3.2511	-3.1109		0.021
-4.6521	-2.0779	-3.2633		0.014
-2.8223	-1.8790	-2.2274		0.027
-0.8921	-0.1650	-0.4954		0.005
-4.6384	-2.5166	-2.2447		0.024
-4.3380	-2.2084	-2.0682		0.016

\* $S = (\sum_i^N (\sigma_i^{\text{exp}} - \sigma_i^{\text{cal}})^2 / N)^{1/2}$ , where  $N$  is the number of experimental data, exp: experimental data and cal: calculated values.

1,4-dimethylbenzene pure components and for the binary systems of dimethyl carbonate + octane, dimethyl carbonate + decane,

diethyl carbonate + octane, diethyl carbonate + decane, dimethyl carbonate + 1,4-dimethylbenzene, diethyl carbonate + 1,4-dimethylbenzene, 1,4-dimethylbenzene + octane, and 1,4-dimethylbenzene + decane at 308.15 K and atmospheric pressure. There is a great amount of information available in the literature<sup>1,9,10,17–38</sup> regarding pure components in a high temperature range; but only a few data<sup>9</sup> are available for same binary mixtures studied in this work. This information is summarized in Table 1.

The surface tension deviation ( $\Delta\sigma$ ), is given by

$$\Delta\sigma / (\text{mN}\cdot\text{m}^{-1}) = \sigma_M - \sum_i x_i \sigma_i \quad (2)$$

where  $\sigma_M$  and  $\sigma_i$  are the surface tension of the mixture and pure component, respectively.

The surface tension deviation  $\Delta\sigma$  was fitted with composition data by the Redlich–Kister polynomial equation,<sup>13</sup> which for binary mixtures is:

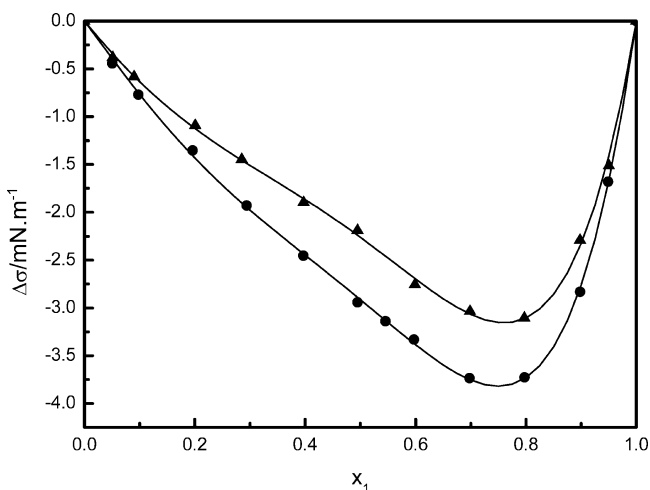
$$\Delta\sigma_{ij} = x_i x_j \sum_k A_k (x_i - x_j)^k \quad (3)$$

where  $x_i$  is the mole fraction of component  $i$ ,  $A_k$  is the polynomial coefficient, and  $k$  is the number of the polynomial coefficient.

Experimental results of surface tension ( $\sigma$ ) and surface tension deviation ( $\Delta\sigma$ ) for the binary mixture at 308.15 are summarized in Table 2.

Table 3 presents the Redlich–Kister coefficients and the standard deviation obtained in the correlations for all binary systems studied at 308.15 K. In each case, the coefficients were obtained by fitting eq 3 by least-squares regression.

Figures 1 to 6 show, as a function of the dialkyl carbonate or 1,4-dimethylbenzene, the surface tension deviation for the



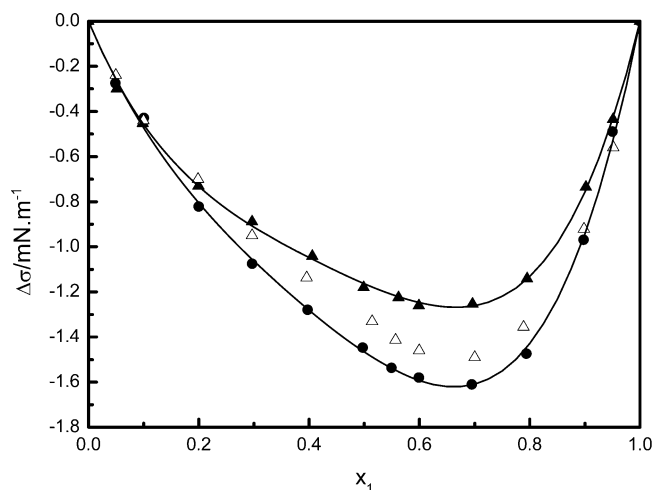
**Figure 1.** Surface tension deviation ( $\Delta\sigma$ ) of dimethyl carbonate (1) + octane (2) (●) and dimethyl carbonate + decane (▲) at 308.15 K. The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

binary mixtures of dialkyl carbonates + alkanes, dialkyl carbonates + 1,4-dimethylbenzene, and 1,4-dimethylbenzene + alkanes at 308.15 K. The symbols correspond to the experimental values, and the continuous lines correspond to the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

**Dialkyl Carbonates + Alkanes Mixtures.** The surface tension deviation ( $\Delta\sigma$ ) is negative for dimethyl carbonate + octane or decane and diethyl carbonate + octane or decane binary mixtures for the entire range of composition and temperature as is shown in Figures 1 and 2, respectively.

Deng et al.<sup>17</sup> have mentioned the surface tension deviations as the result of the surface region aspect and the bulk region aspect. For the surface region, the surface tension deviations represent a nonhomogeneous distribution of molecules between the surface and the bulk of the liquid. Thus, when the concentration of lower surface tension component in the surface is higher than in the bulk, negative surface tension deviations are obtained. On the other hand, in the bulk region chemical and physical effects affect the surface tension deviations. Positive values are obtained when the chemical effects are present, while physical effects and dipolar–dipolar interaction gives negative deviations.<sup>17</sup> In that sense, the physical effects of the dialkyl carbonate + alkanes binary systems are more significant than the chemical effects, which results in a decrease in surface tension.

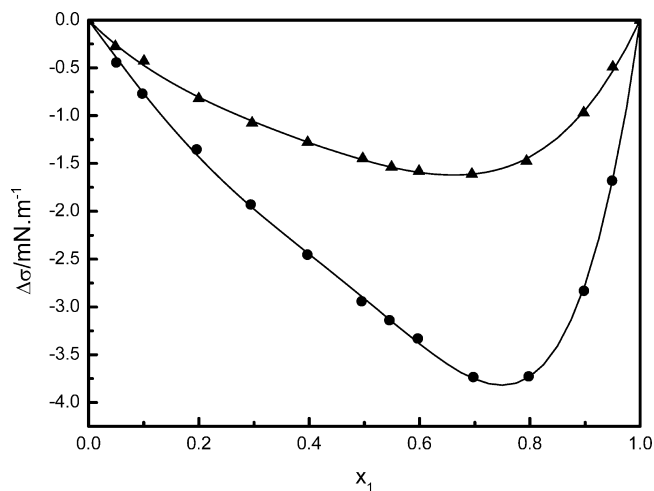
From Figures 1 and 2 an increase of the surface tension deviation in the dialkyl carbonate + alkane with the increasing of the alkane alkyl chain can be seen. Furthermore, Figure 2 shows a comparison between surface tension data measured in this work for diethyl carbonate + decane binary mixtures at 308.15 K with those available in the literature at 298.15 K.<sup>9</sup>



**Figure 2.** Effect of the alkyl chain length of alkane and temperature in the surface tension deviation ( $\Delta\sigma$ ) of diethyl carbonate (1) + octane (2) (●) at 308.15 K and diethyl carbonate + decane at 298.15 K (△) (Mosteiro et al.<sup>9</sup>) and 308.15 K (▲) (this work). The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

From this figure, we can see an increasing of the surface tension deviation with temperature.

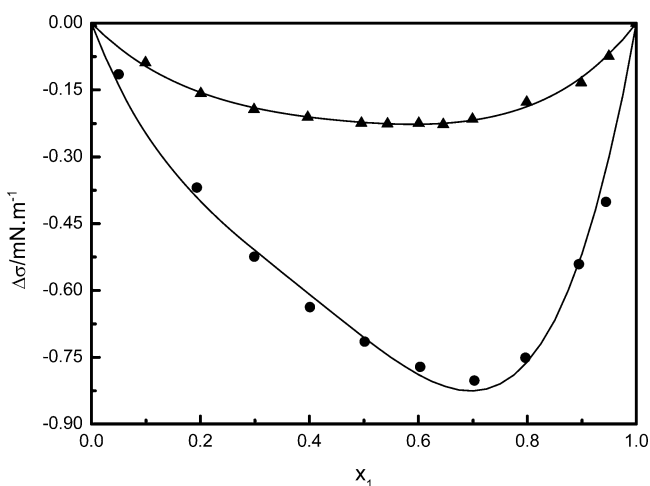
Figure 3 shows a higher surface tension deviation for the diethyl carbonate + octane than the dimethyl carbonate + octane



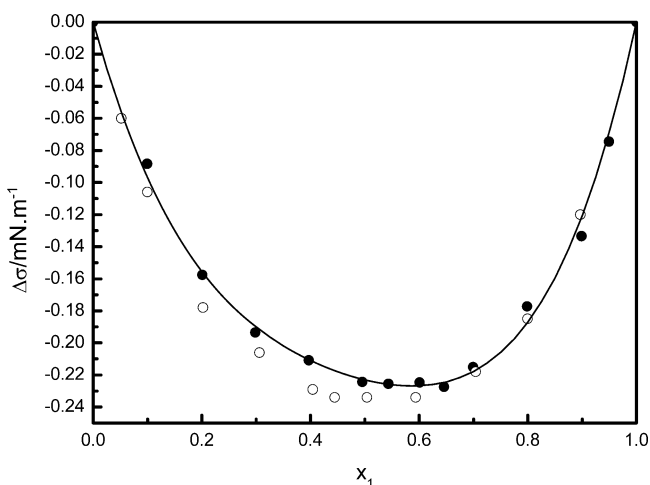
**Figure 3.** Surface tension deviation ( $\Delta\sigma$ ) of dimethyl carbonate (1) + octane (2) (●) and diethyl carbonate (1) + octane (2) (▲) at 308.15 K. The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

binary mixtures. The same behavior was founded for the dialkyl carbonate + decane binary mixtures. The addition of  $\text{CH}_2$  paraffin groups in the alkane or dialkyl carbonate alkyl chains increases the surface tension deviation due to the increasing strength of dispersion forces.<sup>30</sup>

**Dialkyl Carbonates + 1,4-Dimethylbenzene Mixtures.** Again the surface tension deviation ( $\Delta\sigma$ ) is negative for dimethyl carbonate + 1,4-dimethylbenzene and diethyl carbonate + 1,4-dimethylbenzene binary mixtures for the entire range composition and temperature as it can be seen from Figure 4. The diethyl carbonate + 1,4-dimethylbenzene binary mixtures present a higher surface tension deviation than dimethyl



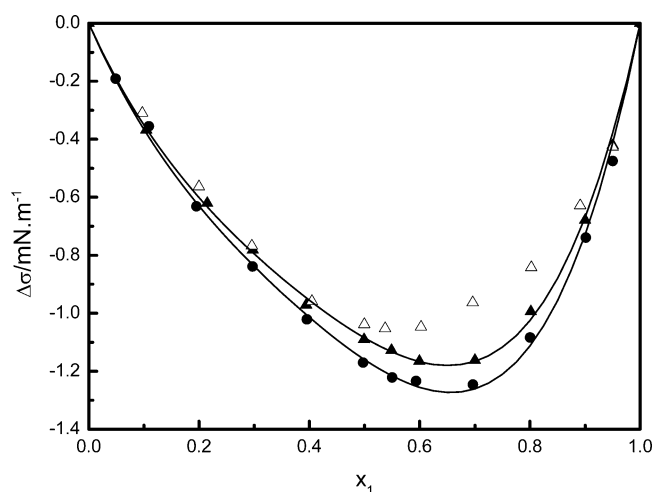
**Figure 4.** Surface tension deviation ( $\Delta\sigma$ ) of dimethyl carbonate (1) + 1,4-dimethylbenzene (2) (●) and diethyl carbonate (1) + 1,4-dimethylbenzene (2) (▲) at 308.15 K. The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.



**Figure 5.** Effect of temperature in surface tension deviation ( $\Delta\sigma$ ) for diethyl carbonate (1) + 1,4-dimethylbenzene (2) at 298.15 K (○, Mosteiro et al.<sup>9</sup>) and 308.15 K (●, this work). The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

carbonate + 1,4-dimethylbenzene. Once more the increasing of the paraffin group  $\text{CH}_2$  in the alkyl chain length increases the surface tension deviation. Figure 5 shows the surface tension deviation obtained in this work for the diethyl carbonate + 1,4-dimethylbenzene at 308.15 K with those reported in the literature at 298.15 K.<sup>9</sup> As for diethyl carbonate + decane binary mixtures, an increase of the surface tension deviation with the temperature can be seen.

**1,4-Dimethylbenzene + Alkanes Mixtures.** Figure 6 shows a negative surface tension deviations at 308.15 K for 1,4-dimethylbenzene + octane and 1,4-dimethylbenzene + decane binary mixtures. The 1,4-dimethylbenzene + decane binary mixtures present a higher surface tension deviation than 1,4-dimethylbenzene + octane binary mixtures because the increasing of the paraffin groups  $\text{CH}_2$  in the alkane alkyl chain increase the surface tension deviation. In addition Figure 6 shows the temperature effect from the 1,4-dimethylbenzene + decane binary mixture at 308.15 K available in this work with those reported in



**Figure 6.** Effect of the alkyl chain length of alkane and temperature in the surface tension deviation ( $\Delta\sigma$ ) of 1,4-dimethylbenzene (1) + octane (2) (●) at 308.15 K and 1,4-dimethylbenzene (1) + decane (2) at 298.15 K (△, Mosteiro et al.<sup>9</sup>) and 308.15 K (▲, this work). The lines are the Redlich–Kister correlations calculated from eq 3 using the parameters listed in Table 3.

the literature at 298.15 K.<sup>9</sup> This figure shows a different behavior with the temperature regarding the systems mentioned before. From this figure an increase of the surface tension deviation can be seen when the temperature decreases. This similar behavior was found for other systems like dimethyl carbonate + 1,2,4-trimethylbenzene or 1,3,5-trimethylbenzene and diethyl carbonate + 1,2,4-trimethylbenzene or 1,3,5-trimethylbenzene binary mixtures.<sup>17</sup>

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### Notes

The authors declare no competing financial interest.

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