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Surface tension and density of the binary mixtures toluene + methyl propanoate, toluene + methyl pentanoate, toluene + methyl heptanoate and toluene + methyl octanoate at atmospheric pressure and 288.15, 298.15 and 308.15 K



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ABSTRACT

Surface tension and density were measured for the binary mixtures toluene + methyl propanoate, toluene + methyl pentanoate, toluene + methyl heptanoate and toluene + methyl octanoate at atmospheric pressure and 288.15, 298.15 and 308.15 K. Meditions were done using mechanical oscillation densimetry and falling drop tensiometry. Densities and surface tensions increases as it grows the carbonated chain of the methyl esters and decreases as temperature increases. Experimental data were satisfactory fitted by the polynomial equation of Redlich-Kister. Adsorption isotherm of Gibbs was applied to estimate the relative adsorption of each ester respect to toluene. The modified group contribution model of UNIFAC was using to estimate the activity coefficients of the components. Positive values of the relative adsorption showed that there was a great accumulation of ester molecules on the surface of the dissolution respect to the fluid's sine.

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Specifications Table

Subject area	<i>Physical Chemistry. Chemical Engineering.</i>
Compounds	<i>Toluene, methyl propanoate, methyl pentanoate, methyl heptanoate, methyl octanoate</i>
Data category	<i>Physicochemical</i>
Data acquisition format	<i>Elemental analysis</i>
Data type	<i>Raw, calculated, analyzed</i>
Procedure	<i>Density was determined with vibrating tube densimeter and surface tension with a falling drop tensiometry. Both properties were measured at different temperatures and atmospheric pressure</i>
Data accessibility	<i>In this article</i>

1. Rationale

The importance that the liquid state has from a chemical point of view is that most chemical processes, which occur in nature, in living organisms, such as those that are conducted in the laboratory and in industry, take place in solution. Liquid state has traditionally been regarded as the most difficult study between the states of matter, which is due largely to the lack of a good model representative of the actual behavior of a liquid system.

The study of liquid mixtures, namely mixtures of liquids with industrial interest, has been a field where, especially in the last few decades, a growing research effort has been invested to meet the scientific and technological needs. This study allows advancing in the knowledge of the molecular interactions through the analysis of the thermodynamic quantities of mixtures. A better understanding of the behavior of the liquid state, facilitate the development of new models for the prediction of thermodynamic properties of binary and multicomponent systems and allows to improving existing ones.

Liquid mixtures formed by aromatic hydrocarbons and esters have industrial importance in its application as components of the solvents for epoxy paints. So, the study of the properties of different combinations of these chemical compounds is of interest to this industry among other chemical industries. One of the lines of our research group conducted during the lasts years, is the measurement of thermo physical properties of mixtures containing aromatic hydrocarbons and linear esters [1–4].

In regards to surface tension, this is a property that reveals information about the surface, the intermolecular interactions and forces in the liquid and exerts a considerable influence on mass and energy transfer through the interface in liquid mixtures. Therefore, the surface tension has a decisive effect in various processes of chemical engineering, such as separation, adsorption, distillation and the performance of biological membranes.

In this case, the research is focused on binary mixtures formed by toluene + methyl esters. Densities and the surface tension of the binary mixtures toluene + methyl propanoate, toluene + methyl pentanoate, toluene + methyl heptanoate, toluene + methyl octanoate were measured at atmospheric pressure and temperatures of 288.15, 298.15 and 308.15 K. Values of density and surface tension were correlated through the polynomial of Redlich–Kister, 1948 [5]. Standard deviations between experimental and calculated values were calculated.

The modified group contribution model of UNIFAC, Gmehling, 1993 [6], was used to predict the activity coefficients of components in the mixtures, necessary to evaluate the relative adsorption by the Gibbs adsorption isotherm.

2. Procedure

The chemicals employed were: Toluene (Sigma-Aldrich, St. Louis, MO, USA, anhydrous, 99.8%, CAS 108-88-3), methyl propanoate (Sigma-Aldrich, St. Louis, MO, USA, 99%, CAS 554-12-1), methyl pentanoate (FlukaChemie AG CH-9470 Buchs, purum 99%, CAS 624-24-8), methyl heptanoate (Fluka-Chemie AG CH-9470 Buchs, Switzerland, puriss > 99%, CAS 106-73-0), methyl octanoate (FlukaChemie AG CH-9470 Buchs, Switzerland, puriss > 99%, CAS 111-11-5). All the liquids were dried over 0.4 nm Union Carbide molecular sieves from Fluka and partially degassed with an ultrasonic bath before use. The mixtures were prepared by weighting. The uncertainty in mole fraction calculation is $\pm 1 \cdot 10^{-4}$.

Table 1

Experimental density, ρ , and surface tension, σ , of pure components contrasted with values found in the literature at 288.15 K, 298.15 K and 308.15 K.

	$\rho/\text{g}\cdot\text{cm}^{-3}$		$\sigma/\text{mN}\cdot\text{m}^{-1}$	
	Exp.	Lit.	Exp.	Lit.
288.15 K				
Toluene	0.87137	0.87200 ^a	29.4	
Methyl propanoate	0.92114	0.92104 ^b	26.3	
Methyl pentanoate	0.89481		26.2	
Methyl heptanoate	0.88482		27.4	
Methyl octanoate	0.88122	0.88150 ^d	28.0	
298.15 K				
Toluene	0.86207	0.86230 ^a	28.2	27.92 ^c
Methyl propanoate	0.90916	0.90903 ^b	24.9	
Methyl pentanoate	0.88475		25.0	
Methyl heptanoate	0.87576	0.87569 ^e	26.3	
Methyl octanoate	0.87254	0.87280 ^d	26.9	
308.15 K				
Toluene	0.85272	0.85400 ^a	26.9	
Methyl propanoate	0.89703	0.89688 ^b	23.4	
Methyl pentanoate	0.87461		24.0	
Methyl heptanoate	0.86666		25.2	
Methyl octanoate	0.86382	0.86400 ^d	25.8	

^a [7].

^b [8].

^c [9].

^d [10].

^e [11].

Densities, ρ , were measured with a digital density and sound velocity analyzer Anton Paar DSA-5000. Calibration was periodically performed using air and double-distilled water. This densimeter has a built-in thermostat which allows working at constant temperature with an error of ± 0.01 K.

Surface tension, σ , was measured using a Lauda TVT2 automated tensiometer with a precision of 10^{-2} $\text{mN}\cdot\text{m}^{-1}$ on the basis of the principle of the drop volume. This technique consists of measuring the volume of a drop detaching from a capillary tube with a circular cross section. The surface tension can be determined as follows:

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

where g is the acceleration of gravity, $\Delta\rho$ is the difference in density between the two phases (liquid or vapor), V is the drop volume, r_{cap} is the internal radius of the capillary and f is a correction factor, defined by the supplier.

In the tensiometer, the syringe and the measuring pail, were thermostated by means of a thermal bath LAUDA PROLINE RP 1840, with a stability of ± 0.01 K. For the determination of the surface tension of systems determined in this work, a syringe of 0.5 mm and a steel capillary radio 1.385 mm were used. The experimental error estimated for the surface tension was ± 0.1 $\text{mN}\cdot\text{m}^{-1}$.

Experimental densities and surface tensions for the binary mixtures were fitted to the polynomial equation of Redlich-Kister [5]:

$$Q = x_1Q_1 + (1 - x_1)Q_2 + x_1(1 - x_1) \sum_{i=0}^p a_i(2x_1 - 1)^i \quad (2)$$

where Q stands for the generic property density or surface tension, x_1 is the mole fraction of component 1. Q_1 and Q_2 are the properties of the pure components, a_i are the adjustment coefficients obtained by least squares and p is the number of coefficients. In each case, standard deviations, s ,

Table 2

Density, ρ , and surface tension, σ , at 288.15 K, 298.15 K and 308.15 K for the binary systems: x_1 Toluene + x_2 Methyl Ester.

x_1	288.15 K		298.15 K		308.15 K	
	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$
x_1 Toluene + x_2 Methyl propanoate						
0.1124	0.91503	26.5	0.90340	25.1	0.89162	23.8
0.1873	0.91114	26.6	0.89972	25.3	0.88819	23.9
0.2747	0.90581	27.1	0.89523	25.7	0.88421	24.3
0.3780	0.90152	27.5	0.89065	25.9	0.87966	24.6
0.4736	0.89662	27.6	0.88601	26.3	0.87528	25.0
0.5824	0.89122	28.2	0.88091	26.7	0.87050	25.4
0.6842	0.88654	28.4	0.87645	27.0	0.86627	25.8
0.7821	0.88165	28.5	0.87183	27.2	0.86193	26.1
0.8849	0.87684	28.8	0.86724	27.5	0.85760	26.3
x_1 Toluene + x_2 Methyl pentanoate						
0.1086	0.89296	26.5	0.88298	25.3	0.87291	24.2
0.1947	0.89135	26.7	0.88143	25.5	0.87143	24.3
0.2865	0.88965	27.0	0.87979	25.7	0.86985	24.5
0.3786	0.88768	27.3	0.87779	26.0	0.86802	24.8
0.4706	0.88572	27.5	0.87599	26.3	0.86619	25.1
0.5882	0.88302	27.9	0.87337	26.6	0.86367	25.4
0.6977	0.88027	28.3	0.87076	26.9	0.86114	25.7
0.7955	0.87759	28.5	0.86812	27.2	0.85859	26.0
0.8941	0.87461	28.9	0.86522	27.6	0.85576	26.4
x_1 Toluene + x_2 Methyl heptanoate						
0.1028	0.88416	27.6	0.87508	26.5	0.86595	25.4
0.1959	0.88345	27.8	0.87436	26.6	0.86522	25.5
0.2917	0.88257	27.9	0.87347	26.7	0.86431	25.6
0.3862	0.88162	28.0	0.87250	26.8	0.86333	25.8
0.4820	0.88053	28.2	0.87140	27.0	0.86221	25.9
0.5923	0.87912	28.4	0.86995	27.2	0.86074	26.1
0.7017	0.87748	28.5	0.86831	27.4	0.85904	26.2
0.7996	0.87576	28.8	0.86655	27.5	0.85727	26.4
0.8958	0.87382	29.0	0.86457	27.7	0.85527	26.5
x_1 Toluene + x_2 Methyl octanoate						
0.1125	0.88074	28.1	0.87202	27.0	0.86326	25.9
0.1962	0.88031	28.2	0.87156	27.0	0.86278	25.9
0.2897	0.87984	28.2	0.87106	27.1	0.86223	26.0
0.3872	0.87913	28.4	0.87031	27.2	0.86146	26.1
0.4947	0.87832	28.5	0.86944	27.3	0.86051	26.2
0.6071	0.87728	28.7	0.86834	27.4	0.85935	26.3
0.7055	0.87616	28.7	0.86715	27.5	0.85810	26.4
0.7994	0.87491	29.1	0.86582	27.8	0.85669	26.6
0.8941	0.87335	29.3	0.86418	28.0	0.85495	26.8

were calculated as follows:

$$s = \left[\sum_{i=1}^n (Q_{exp} - Q_{calc})^2 / (n - p) \right]^{1/2} \quad (3)$$

where Q stand for density or surface tension of the binary mixtures, n is the number of data and p the number of coefficients.

The experimental data of surface tension were also used to applied the well know Gibbs isotherm. The model defines surface excess concentration as the ratio between the number of moles in excess of component i . per unit area:

$$I_i^E = \frac{n_i^E}{A^E} \quad (4)$$

Table 3

Coefficients, a_i , and standard deviation, s , for density and surface tension of the binary systems at 288.15 K, 298.15 K and 308.15 K, Equation (3).

	288.15 K		298.15 K		308.15 K	
	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$	$\rho/\text{g}\cdot\text{cm}^{-3}$	$\sigma/\text{mN}\cdot\text{m}^{-1}$
x_1 Toluene + x_2 Methyl propanoate						
a_0	-0.00360	-0.48	-0.00321	-0.81	-0.00099	-0.35
a_1	0.00124	-0.05	0.00112	-0.57	0.00131	0.01
a_2	-	-	-	-	-	-
s	0.00008	0.12	0.00007	0.11	0.00007	0.07
x_1 Toluene + x_2 Methyl pentanoate						
a_0	0.00796	-0.85	0.00789	-1.15	0.00776	-0.95
a_1	0.00122	-0.08	0.00133	-0.99	0.00131	-0.20
a_2	-	-0.53	-	-1.68	-	-1.06
s	0.00003	0.02	0.00003	0.03	0.00003	0.02
x_1 Toluene + x_2 Methyl heptanoate						
a_0	0.00889	-0.97	0.00906	-0.79	0.00917	-0.29
a_1	0.00223	-0.85	0.00235	-1.35	0.00241	-0.92
a_2	0.00109	-	0.00114	-0.08	0.00105	-
s	0.00001	0.04	0.00001	0.05	0.00001	0.03
x_1 Toluene + x_2 Methyl octanoate						
a_0	0.00795	-0.92	0.00840	-1.05	0.00885	-0.56
a_1	0.00253	-0.02	0.00271	-0.25	0.00287	-0.05
a_2	0.00045	-	0.00048	-	0.00053	-
s	0.00002	0.04	0.00002	0.03	0.00002	0.02

It is a measure of a specie accumulation in the interface due to adsorption. As n_i^s and therefore Γ_i , depend on the arbitrary location given to the splitter plane. It is possible to choose a position for the plane where $\Gamma_i = 0$. Under these conditions, Gibbs called to the surface excess of solute concentration, adsorption. Since σ , surface tension of a liquid, measures the free energy per unit area of the liquid-air interface, there will be an enrichment at the interface (respect to the inside of the solution) of the component that decreases the surface tension; to make the system free energy a minimum. If solute 2 decreases the surface tension of the solvent 1, the surface layer will be more concentrated than the interior of the solution. An opposite case arises when aggregating 2 to 1 causes an increase in the surface tension. It is said that a positive or negative solute adsorption takes place respectively.

If we assume that there is no adsorption for the solvent, Eq. (4) is derived to:

$$\Gamma_{2(1)} = -\frac{1}{RT} \frac{d\sigma}{d\ln(a_2)} \quad (5)$$

where $\Gamma_{2(1)}$ is the relative adsorption of component 2 with respect to component 1 and $a_2 = \gamma_2 x_2$. Units of $\Gamma_{2(1)}$ are mol m^{-2} . It can be observed from Eq. (5), that when the concentration of solute increases with the surface tension of the solution, the ratio will be positive and $\Gamma_{2(1)}$ will be negative, and the concentration on the surface will be lower than in the solution. The Gibbs isotherm was applied to four binary systems formed by toluene and esters. We accomplished the calculation of the relative adsorption, $\Gamma_{2(1)}$, taking toluene as component 1, which is the component common to all systems, and each ester as component 2. To apply Eq. (5) it is necessary to know the activity coefficient, γ_2 . Activity coefficients were estimated using the modified group contribution model of UNIFAC [6].

3. Data, value and validation

The measured properties of these chemicals compared to those found in literature are presented in Table 1.

Experimental densities and surface tensions for the binary mixtures at 288.15 K, 298.15 K and 308.15 K are reported in Table 2. In Table 3, coefficients a_i for Eq. (2) and standard deviations, s , for the density and surface tension of the binary mixtures are listed at the three temperatures studied.

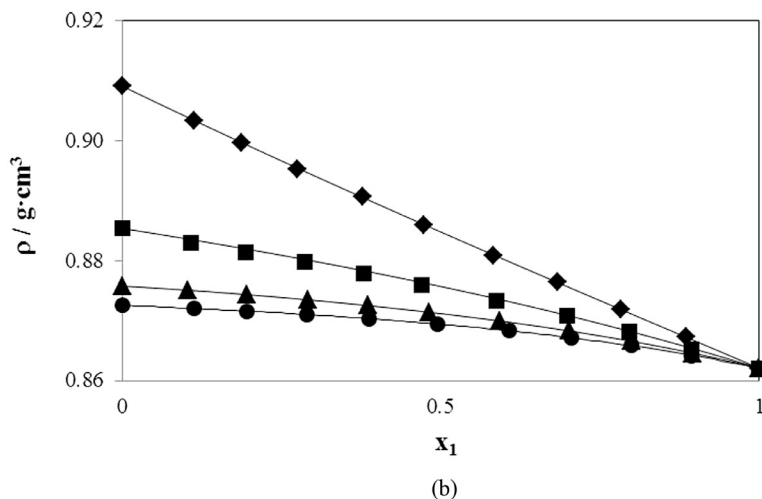
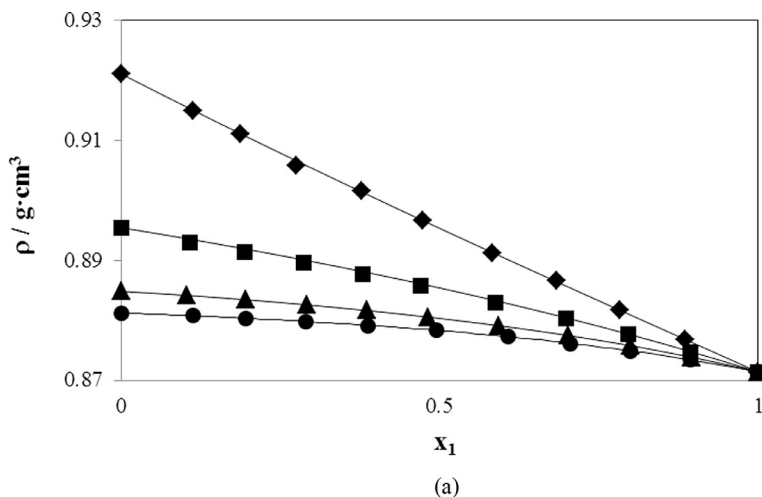


Fig. 1. Density, ρ , vs x_1 for the binary systems: x_1 Toluene + x_2 Methyl propanoate, (\blacklozenge); x_1 Toluene + x_2 Methyl pentanoate, (\blacksquare), x_1 Toluene + x_2 Methyl heptanoate, (\blacktriangle); x_1 Toluene + x_2 Methyl octanoate, (\bullet) at (a) 288.15, (b) 298.15, (c) 308.15 K. Continuous curves were calculated with polynomial Eq. (2).

Fig. 1(a)–(c) depict density vs mole fraction of toluene of the systems studied at 288.15 K, 298.15 K and 308.15 K, respectively. Densities increase as it grows the carbonated chain of the methyl esters and decrease as temperature increases.

Fig. 2(a)–(c) show that the surface tension of the systems studied increases as it grows the carbonated chain of the methyl esters. It can be observed that surface tension decreases gradually with increasing concentration of the esters. Besides, an increase in temperature produces a marked decrease in the surface tension of mixtures and pure components, as can be seen from the analysis of these figures.

Table 4 includes the activity coefficient, γ_2 , estimated with the modified group contribution model of UNIFAC [6] at 298.15 K, and used for the surface excess concentration calculations.

The adsorption process for the systems considered was evaluated by calculating the relative adsorption, $\Gamma_{2(1)}$, by the Gibbs isotherm (Eq. (5)). As it can be observed from Fig. 3, the four binary systems exhibited positive values of $\Gamma_{2(1)}$ at 298.15 K. This is indicative of a preferential adsorption at

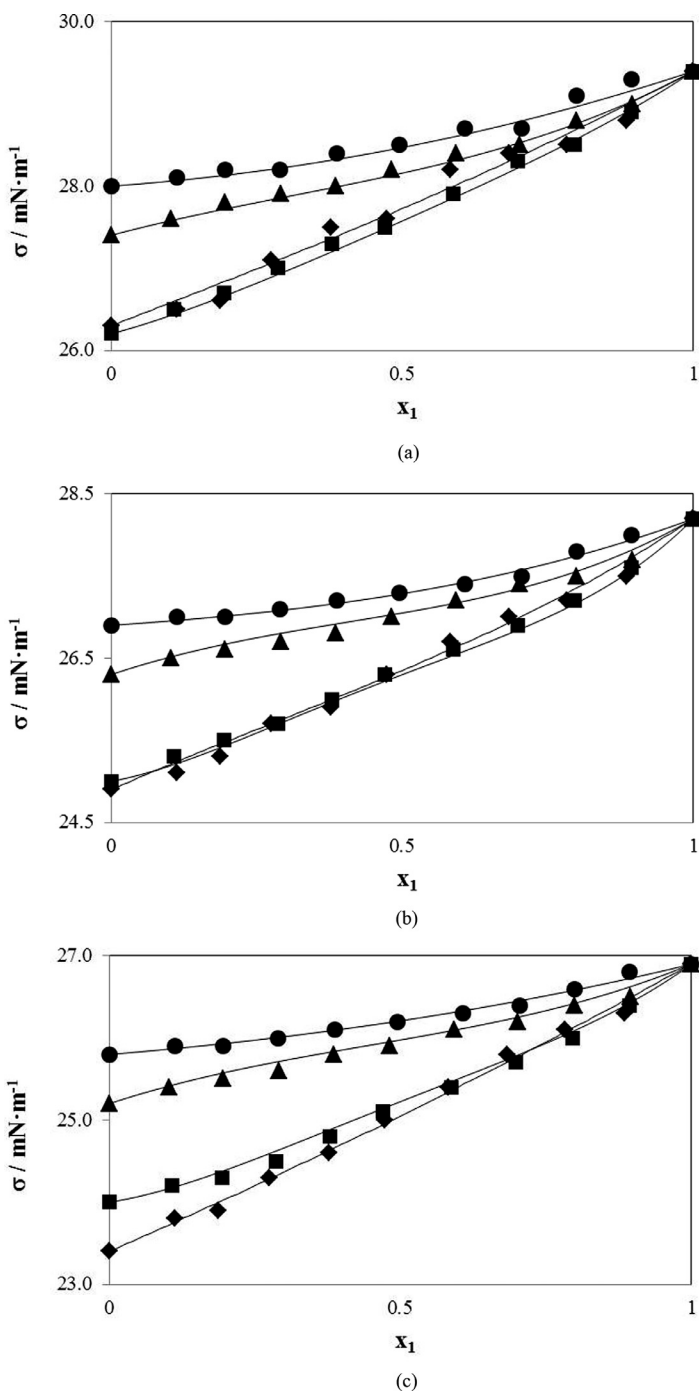


Fig. 2. Surface tension, σ , vs x_1 for the binary systems: x_1 Toluene + x_2 Methyl propanoate, (\blacklozenge); x_1 Toluene + x_2 Methyl pentanoate, (\blacksquare), x_1 Toluene + x_2 Methyl heptanoate, (\blacktriangle); x_1 Toluene + x_2 Methyl octanoate, (\bullet) at (a) 288.15, (b) 298.15, (c) 308.15 K. Continuous curves were calculated with polynomial Eq. (2).

Table 4

Activity coefficient, γ_2 , estimated with the modified group contribution model of UNIFAC [6] at 298.15 K for the binary systems: x_1 Toluene + x_2 Methyl Ester.

x_2	γ_2	x_2	γ_2	x_2	γ_2
x_1 Toluene + x_2 Methyl propanoate					
0.1151	1.3340	0.4152	1.1413	0.7253	1.0308
0.2138	1.2605	0.5254	1.0924	0.8080	1.0150
0.3158	1.1952	0.6105	1.0620	0.9042	1.0038
x_1 Toluene + x_2 Methyl pentanoate					
0.1059	1.0540	0.4118	1.0147	0.7135	1.0010
0.2045	1.0387	0.5294	1.0068	0.8053	1.0001
0.3023	1.0447	0.6214	1.0031	0.8914	0.9999
x_1 Toluene + x_2 Methyl heptanoate					
0.1042	0.9029	0.4077	0.9578	0.7083	0.9889
0.2004	0.9251	0.5180	0.9712	0.8041	0.9949
0.2983	0.9422	0.6138	0.9810	0.8972	0.9986
x_1 Toluene + x_2 Methyl octanoate					
0.1059	0.8508	0.3929	0.9367	0.7103	0.9858
0.2006	0.8867	0.5053	0.9582	0.8038	0.9935
0.2945	0.9137	0.6128	0.9745	0.8875	0.9979

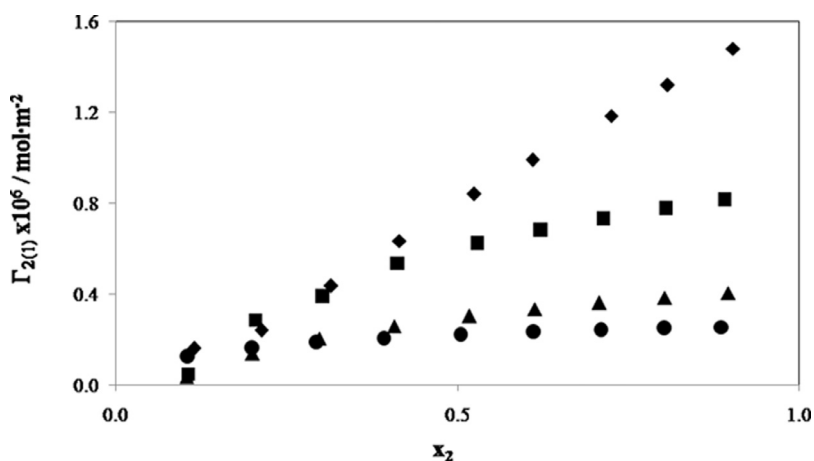


Fig. 3. Relative adsorption of component 2 with respect to component 1, $\Gamma_{2(1)}$, vs the mole fraction of component 2 at 298,15 K for the binary systems: x_1 Toluene + x_2 Methyl propanoate, (\blacklozenge); x_1 Toluene + x_2 Methyl pentanoate, (\blacksquare); x_1 Toluene + x_2 Methyl heptanoate, (\blacktriangle); x_1 Toluene + x_2 Methyl octanoate, (\bullet).

the interface of the methyl ester (2), either methyl propanoate, methyl pentanoate, methyl heptanoate or methyl octanoate, over the toluene (1). Qualitatively, in all cases an approximate linear increase of $\Gamma_{2(1)}$ as a function of x_2 is observed.

As all systems have a positive adsorption of the methyl ester, it is expected that the addition of this component to the mixture produces a decrease in the surface tension of it. Consequently, the behavior of the relative adsorption with the composition is inverse to that observed for the surface tension. In addition, these values are significantly higher for the binary system formed from methyl propanoate than for that formed by methyl octanoate. This allows to conclude that the tendency to preferentially migrate to the interface is smaller as the length of the carbon chain of the methyl ester increases.

4. Conclusions

Experimental determination of density and surface tension of the binary mixtures toluene + methyl propanoate, toluene + methyl pentanoate, toluene + methyl heptanoate and toluene + methyl octanoate at atmospheric pressure and 288.15, 298.15 and 308.15 K was performed using mechanical oscillation densimetry and falling drop tensiometry. The measured properties exhibited a decreasing tendency towards an increase in the temperature of the system, which corresponds to the habitual behavior observed for the liquid systems.

The experimental and calculated values were satisfactory correlated through the Redlich-Kister's polynomial equation. Additionally, the adsorption isotherm of Gibbs was applied to analyze the relative adsorption of one component respect to the other in the binary system, using the modified group contribution model of UNIFAC to estimate the activity coefficients of the components. The application of the Gibbs isotherm allowed to conclude that for both systems there was a superficial excess of the methyl ester, indicating that this solute tends to adsorb at the liquid - vapor interface. This is consistent with the fact that esters have lower surface tensions than toluene.

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