

Measuring the Density, Viscosity, Surface Tension, and Refractive Index of Binary Mixtures of Cetane with Solketal, a Novel Fuel Additive

Jesús Esteban,^{*,†} Halina Murasiewicz,^{†,‡} Tom A. H. Simons,[†] Serafim Bakalis,[†] and Peter J. Fryer[†]

[†]School of Chemical Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

[‡]West Pomeranian University of Technology, Aleja Piastów 17, 70-310 Szczecin, Poland

ABSTRACT: As a consequence of the surplus of glycerol generated from the development of the biodiesel industry, making valuable products from it has been an ongoing subject for the past few years. In previous work, several products have been synthesized from glycerol, such as solketal, with a proven value as an additive to different types of fuels. On the other hand, *n*-hexadecane is frequently used as a reference compound to model diesel properties. This work presents a thorough experimental study on the determination of different volumetric properties of binary mixtures containing *n*-hexadecane and solketal, namely, density, viscosity, surface tension, refractive index, and the excess properties and deviations thereof derived. These properties were measured over the entire range of compositions in increments of the molar fraction of the components of 0.1 in the range from 293.15 or 298.15 to 323.15 K. Finally, correlation of the Redlich–Kister equation to the experimental excess properties and deviations was made, allowing for the retrieval of the corresponding fitting parameters.

1. INTRODUCTION

It is well-known that one of the grand challenges of the biodiesel industry is to manage the oversupply of glycerol generated as a byproduct of its manufacturing process. The surplus of glycerol has eventually led to a significant drop of its price, which can damage the overall economy of the process.¹ One way to exploit the surplus of glycerol has been to conduct studies on its chemical valorization to yield value-added products of interest in several applications.^{2,3} Examples would be the production of glycerol carbonate^{4,5} or esters.^{6–8}

Of interest to the fuel and energy science, oxygenate compounds have been synthesized from glycerol, such as ethers^{9–11} or acetals,^{12–15} which have proven effective as fuel additives. To the latter family of compounds belongs solketal, which has demonstrated to enhance performance parameters of gasoline, biodiesel, and diesel alike.^{16–18}

In addition, *n*-hexadecane, also known as cetane, is a reference compound commonly used as a model diesel molecule. In fact, the so-called cetane number is a key indicator of the performance of a diesel fuel and refers to a measure of its ignition delay within an engine. Studies making use of *n*-hexadecane as model diesel include studies of the removal of nitrogen compounds with deep eutectic solvents or ionic liquids,^{19,20} the extraction of benzene, toluene, and xylenes (BTX) aromatics,²¹ or the addition of different methylesters as biodiesel additives.²²

Understanding the volumetric and transport properties of fuels is of crucial significance to the design of engines, because they have an effect on the combustion quality; in addition, good knowledge of these properties and the deviations observed are important for a more accurate design of industrial equipment.^{23–26} Internal combustion engines require atomization of the fuel before combustion, which is influenced by many factors of the fuel itself, including density, viscosity, and surface tension.^{27–29} The addition of oxygenate compounds has proven

to reduce surface tension and viscosity of blends,^{30,31} which in the end will enhance atomization of the fuel.

As a result, the determination of volumetric, transport, and refractive properties has been undertaken for several mixtures containing *n*-hexadecane.^{22,32–39}

In this work, we aim at filling a gap found in the literature concerning the use of solketal as a diesel additive. Thus, the density, viscosity, surface tension, and refractive index of the binary mixtures of *n*-hexadecane and solketal have been measured over the entire composition range at temperatures ranging from 293.15 to 323.15 K. In addition, excess properties and deviations have been calculated from the experimental data, and fitting of the Redlich–Kister equation was made to such values.

2. MATERIALS AND METHODS

2.1. Materials and Preparation of the Binary Mixtures. *n*-Hexadecane (purity of >99%) and solketal (DL-1,2-isopropylidene-glycerol, purity of >98%) samples were supplied by Sigma-Aldrich Co., Ltd. Both chemicals were used as received from the suppliers.

The corresponding binary mixtures were prepared by weighing in a XB 220A analytical balance (Precisa Gravimetrics, AG, Switzerland) with ± 0.0001 g accuracy. As well as measuring the properties of the pure compounds, nine binary mixtures were made with the intention of varying the molar fraction of *n*-hexadecane and solketal in intervals of 0.1.

2.2. Density Measurements. The density of the materials was determined with a tensiometer (Krüss K100, Germany) in measurements based on the Archimedes principle. First, to tare the tensiometer balance, an inert platinum–iridium bident-shaped hook holder (16 mm from dent to dent) was immersed in a cylindrical vessel containing 80 mL of the liquid sample to be analyzed.

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Table 1. Comparison of Experimental Density (ρ), Viscosity (η), Surface Tension (γ), and Refractive Index (n_D) for Pure *n*-Hexadecane and Solketal at Different Temperatures

component	<i>T</i> (K)	ρ (g cm ⁻³)		μ (mPa s)		γ (mN m ⁻¹)		n_D	
		this work	literature	this work	literature	this work	literature	this work	literature
<i>n</i> -hexadecane	293.15			3.384	3.447 ⁴⁴				
	298.15	0.7701	0.7702 ⁴¹ 0.7699 ⁴⁴ 0.7733 ⁴² 0.7706 ⁴³ 0.76992 ²²	3.070	3.065 ⁴¹ 3.061 ⁴⁴ 3.039 ⁴³ 3.041 ⁴⁰ 3.056 ²²	27.3268		1.43467	1.43244 ⁴¹ 1.4348 ⁴² 1.4340 ⁴³
	303.15	0.7670	0.766981 ⁴⁰ 0.76651 ²²	2.756	2.706 ⁴⁰ 2.727 ²²	27.0201	27.05 ³⁹	1.43228	
	313.15	0.7600	0.7598 ⁴¹ 0.7597 ⁴⁴	2.226	2.219 ⁴¹ 2.458 ⁴⁴	26.2202	26.26 ³⁹	1.42997	
	323.15	0.7540	0.75268 ²²	1.895	1.837 ²²	25.3231	25.30 ³⁹	1.42795	
	298.15	1.0629	1.055 ^{a,46}	9.643	11 ⁴⁵	32.0788	32.10 ^{b,45}	1.43360	1.4326 ^{a,46} 1.4325 ^{c,47} 1.4309 ^{b,45}
	298.15		1.0624 ^{c,47}						
	298.15		1.066 ^{b,45}						
solketal	298.15	1.0629	1.055 ^{a,46}	9.643	11 ⁴⁵	32.0788	32.10 ^{b,45}	1.43360	1.4326 ^{a,46} 1.4325 ^{c,47} 1.4309 ^{b,45}
	298.15		1.0624 ^{c,47}						
	298.15		1.066 ^{b,45}						

^aThe reported purity is 94.4%. ^bThe reported purity is 96.5%. ^cThe reported purity is 84%.

Afterward, a silicon crystal density standard (2.3 g cm⁻³) placed on the hook holder was immersed, and measurements were recorded. The accuracy of the measurements was ± 0.0001 g cm⁻³. The vessel containing the liquid sample was placed on a jacketed vessel connected to a temperature-controlled water bath (Tecam TE-7 Tempette, Italy), with which the temperature could be controlled with an accuracy of ± 0.01 K. Three repetitions of each measurement were completed. The expanded uncertainty ($k = 2$) of the measurements was estimated to be below 0.0003 g cm⁻³ in all cases.

2.3. Viscosity Measurements. The viscosities of pure components of *n*-hexadecane and solketal and of binary mixtures were measured using a Bohlin rheometer (Malvern Instruments, Ltd., U.K.) fitted with cone and plate geometry, 2°/55 mm. The temperature was supplied and controlled through the plate with an uncertainty of ± 0.01 K. After determination that the samples showed Newtonian behavior (data not shown), the applied shear rate was fixed at 200 s⁻¹. The dynamic viscosities were determined from the corresponding flow curves with an accuracy of ± 0.001 mPa s. Triplicate measurements of each sample were realized, with the expanded uncertainty ($k = 2$) of the viscosity measurements being calculated to be 0.002 mPa s at most.

2.4. Surface Tension Measurements. The surface tension of the materials was also determined making use of a tensiometer (Krüss K100, Germany) with the same sample vessel and temperature control system as described in section 2.2. On this occasion, a platinum Wilhelmy plate geometry of 20 mm of length and a thickness of 0.18 mm was employed to such effect.

The initial immersion of the plate into the sample was 2.00 mm, after which the device was programmed to take 50 measurements over a time frame of 1000 s or less if the surface tension was determined constant (a variation of the standard deviation of less than 0.01 for 10 consecutive measurements). Again, triplicate experiments were performed. Surface tensions with the Wilhelmy plate method could be determined with a resolution of ± 0.0002 mN m⁻¹, and the expanded uncertainty ($k = 2$) of these observations was below 0.0005 mN m⁻¹.

2.5. Refractive Index Measurements. The refractive indices of the samples were measured with a Rudolph J357 refractometer (Rudolph Research Analytical, Hackettstown, NJ) controlling temperature (with a reproducibility of ± 0.01 K). The accuracy in the readings of the refractive index instrument equaled ± 0.0002 . Before the measurement of each sample, the refractometer was calibrated with deionized water at 293.15 K. The value of the refractive index for each sample was calculated as an average obtained from three single measurements. In this case, the expanded uncertainty ($k = 2$) was not above 0.0005.

3. RESULTS AND CORRELATIONS

Validation of the experimental values obtained of each of the properties with the equipment employed in this work was made by comparison to literature values that exist in the literature for *n*-hexadecane^{22,39–44} and solketal.^{45–47} Table 1 compiles values reported in the literature and our experimental observations at different temperatures. It is worthwhile remarking the significant difference in the amount of experimental data found for each of the chemicals. While there is considerable experimental data on pure *n*-hexadecane, especially concerning densities and viscosities, owing to its traditional use as a model compound for diesel, there is very limited information about solketal. For this compound, the very limited data existing in the literature refer to the product with a lower degree of purity to that reported herein, which may be the case why a relative error of 12.3% was found when comparing its viscosity at 25 °C to that reported for a commercial sample. With that sole exception, it can be observed that the deviations of each of the properties are very low, with the relative deviation of the viscosity of *n*-hexadecane at 20 °C at 1.83% being the highest of them.

3.1. Volumetric Properties. Table 2 summarizes the experimental values of density acquired for the pure compounds as well as the nine binary mixtures of increasing molar fractions of 0.1 at 298.15, 303.15, 313.15, and 323.15 K and $P = 0.1$ MPa. In addition, the excess molar volumes V^E of the binary mixtures were computed according to the following equation:

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \quad (1)$$

in which the subscripts 1 and 2 are for *n*-hexadecane and solketal, respectively, with x being the molar fraction, M being the molar weight, and ρ being the density of both of the mixtures (without subscript) and the pure components.

Figure 1 depicts the dependence of the molar excess volume of the binary mixtures of cetane and solketal with respect to their molarity at different temperatures. The figure features both the experimental values and the fitting to the Redlich–Kister equation,⁴⁸ given by the following equation:

Table 2. Densities (ρ) and Excess Molar Volumes (V^E) for the Binary Mixtures Consisting of *n*-Hexadecane (1) + Solketal (2) at 298.15, 303.15, 313.15, and 323.15 K

x_1	ρ (g cm ⁻³)	V^E (cm ³ mol ⁻¹)	ρ (g cm ⁻³)	V^E (cm ³ mol ⁻¹)
<i>T</i> = 298.15 K				
0.0000	1.0629	0.0000	1.0501	0.0000
0.1016	1.0618	-8.0885	1.0364	-6.5887
0.2001	1.0237	-10.7752	0.9971	-8.8245
0.2991	0.9375	-4.0558	0.9198	-2.9982
0.4008	0.8486	7.9059	0.8337	9.0227
0.5000	0.8069	13.0112	0.7926	14.3058
0.5992	0.7850	14.2967	0.7694	16.2189
0.7069	0.7782	11.1573	0.7661	11.9979
0.7999	0.7772	7.0063	0.7644	8.0565
0.8998	0.7790	1.5008	0.7650	2.9408
1.0000	0.7701	0.0000	0.7600	0.0000
<i>T</i> = 303.15 K				
0.0000	1.0580	0.0000	1.0420	0.0000
0.1016	1.0522	-7.5112	1.0170	-5.0896
0.2001	1.0094	-9.3768	0.9833	-7.9589
0.2991	0.9291	-3.2665	0.9070	-1.9221
0.4008	0.8434	8.3328	0.8265	9.2672
0.5000	0.8015	13.6228	0.7841	15.0899
0.5992	0.7783	15.4226	0.7618	16.8539
0.7069	0.7753	11.1173	0.7578	12.8752
0.7999	0.7736	7.1740	0.7574	8.4505
0.8998	0.7742	2.1113	0.7589	3.0014
1.0000	0.7670	0.0000	0.7540	0.0000
<i>T</i> = 313.15 K				
0.0000	1.0580	0.0000	1.0420	0.0000
0.1016	1.0522	-7.5112	1.0170	-5.0896
0.2001	1.0094	-9.3768	0.9833	-7.9589
0.2991	0.9291	-3.2665	0.9070	-1.9221
0.4008	0.8434	8.3328	0.8265	9.2672
0.5000	0.8015	13.6228	0.7841	15.0899
0.5992	0.7783	15.4226	0.7618	16.8539
0.7069	0.7753	11.1173	0.7578	12.8752
0.7999	0.7736	7.1740	0.7574	8.4505
0.8998	0.7742	2.1113	0.7589	3.0014
1.0000	0.7670	0.0000	0.7540	0.0000

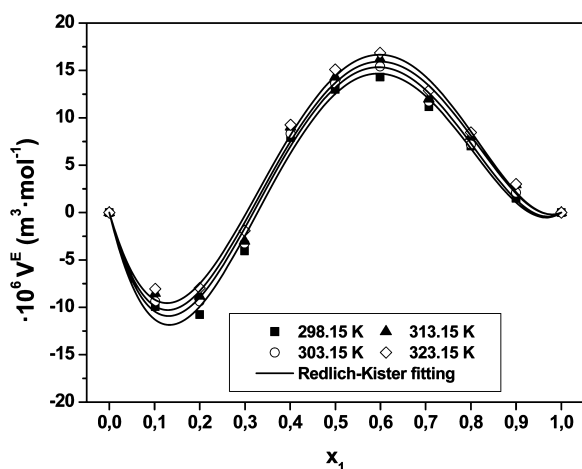


Figure 1. Excess molar volumes for the binary mixture of *n*-hexadecane (1) + solketal (2) at 298.15, 303.15, 313.15, and 323.15 K. Points show experimental observations, and the solid curve shows the Redlich–Kister fitting.

$$P = x_1(1 - x_1) \sum_{i=0}^n A_i(1 - x_1)^i \quad (2)$$

where P stands for the molar excess volume in this case, A_i are the fitting parameters of the equation, and n is the number of parameters present in the fitting. Equation 2 includes the term P in a generic form as a result of the fact that it is also used for fitting the deviation of other properties, as will be discussed in subsequent sections.

Commonly, eq 2 features from four⁴⁹ to six²² parameters; herein, the fitting has been performed making use of a total of four parameters. The values of the correlation parameters can

be found in Table 3 together with the standard deviation (σ), computed with eq 3

$$\sigma = \sqrt{\frac{\sum (Y^E - Y_{\text{cal}}^E)}{(N - n)}} \quad (3)$$

where Y^E stands for the observed value of the molar excess volume or property deviation and Y_{cal}^E stands for said property calculated from the Redlich–Kister correlation. In addition, N_{exp} is the number of experimental data points, in this case 11, and n is the number of parameters, in this case 4, as discussed.

With regard to the dependence of V^E with respect to the composition of the binary mixtures, represented by the molar composition of cetane x_1 , it can be observed in Figure 1 that the curve (i) initially acquires negative values for molar fractions of cetane up to 0.3 and then (ii) switches to positive values. This behavior has previously been observed in mixtures of 2,2,4-trimethylpentane with 1-alcohols.⁵⁰ Considering the similar nature of the compounds dealt with herein (cetane is an alkane, and solketal is an alcohol), these deviations can be ascribed to different contributions. The positive V^E values are thought to be caused by packing effects in the mixture, with an increasing presence of cetane leading to the disruption of the hydrogen bonding that may form among solketal molecules. The negative values, for their part, may be caused by packing of oversized molecules, which can result in interstitial void space that becomes filled by smaller molecules.⁵¹ As for the effect of the temperature, an increase causes the hydrogen bonds to break, which is the reason why the values are more and more positive as this variable increases.⁵²

3.2. Viscometric Properties. Experimental dynamic viscosities of the same binary mixtures were determined at 293.15, 303.15, 313.15, and 323.15 K at $P = 0.1$ MPa. The values are presented in Table 4 together with the viscosity deviations, which were calculated with the expression

$$\Delta\mu = \mu - (x_1\mu_1 + (1 - x_1)\mu_2) \quad (4)$$

where μ is the absolute dynamic viscosity of the mixture and μ_1 and μ_2 are those of the pure components *n*-hexadecane and solketal, respectively. Furthermore, Table 4 also includes the excess Gibbs free energy of activation for viscous flow, $\Delta G^{\text{E}51}$

$$\Delta G^{\text{E}} = RT[\ln(\mu V) - (x_1 \ln(\mu_1 V_1) + (1 - x_1) \ln(\mu_2 V_2))] \quad (5)$$

in which R is the ideal gas constant, T is the temperature, and V , V_1 , and V_2 refer to the molar volumes of the mixture and pure compounds. V can be calculated as

$$V = \frac{x_1 M_1 + x_2 M_2}{\rho} \quad (6)$$

that is, the first addend of the definition of the molar excess volume according to the definition given in eq 1.

Figures 2 and 3 show the dependence of $\Delta\mu$ and ΔG^{E} with respect to the molar fraction of each binary mixture at the temperatures tested including experimental values and the fitting to the Redlich–Kister equation. The corresponding parameters retrieved from correlation are featured in Table 3.

The evolution of the viscosity deviation with an increasing concentration of cetane follows a set of W-shaped curves for each temperature, which consists of two regions of negative values separated by one region acquiring positive values. Qualitatively, identical behavior has been identified in the case

Table 3. Parameters Retrieved and Standard Deviations Calculated from Fitting the Redlich–Kister Equation to the Observed Excess Molar Volumes and Deviations of Viscosity, Surface Tension, and Refractive Index

excess or deviation property	T (K)	A ₀	A ₁	A ₂	A ₃	σ
V ^E	298.15	−32.8335	518.0833	−722.9178	37.2144	1.5812
	303.15	−29.7632	525.1761	−758.0131	75.8892	1.5302
	313.15	−20.3290	499.3544	−739.5356	83.0081	1.7143
	323.15	−20.8681	528.4170	−812.3382	139.0596	1.8310
Δμ	293.15	−11.8755	70.6208	−52.5023	−32.4753	0.0637
	303.15	−5.2976	19.0447	36.2925	−71.1248	0.0254
	313.15	−1.7909	−12.7615	93.9955	−97.1384	0.0557
	323.15	−3.2623	−7.7877	73.5994	−74.1029	0.0974
ΔG ^E	293.15	−1511.1589	20107.4671	−29413.4581	8417.7356	25.2947
	303.15	−968.0023	13374.3496	−13346.0155	−2066.3343	27.7991
	313.15	−434.5701	4361.3664	8858.6035	−16631.8394	22.1469
	323.15	−1963.6788	9759.3671	270.7819	−10941.7145	43.9191
Δγ	298.15	−50.5994	143.5303	−212.5439	59.5026	0.0712
	303.15	−49.0329	142.6095	−216.6726	65.9997	0.1508
	313.15	−43.3390	117.6239	−178.4413	50.4304	0.0857
	323.15	−41.7275	117.4966	−183.7589	56.7663	0.1010
ΔR _m	293.15	10.8585	−87.9727	368.7946	−351.3428	1.6130
	303.15	13.7793	−109.1836	421.6168	−386.6239	1.7634
	313.15	10.8585	−87.9727	368.7946	−351.3428	1.5726
	323.15	10.2474	−63.7777	312.3705	−309.0096	1.7400

Table 4. Dynamic Viscosities (μ), Viscosity Deviations (Δμ), and Excess Gibbs Free Energy of Activation of Viscous Flow (ΔG^E) for the Binary Mixtures of *n*-Hexadecane (1) + Solketal (2) at 293.15, 303.15, 313.15, and 323.15 K

x ₁	μ (mPa s)	Δμ (mPa s)	ΔG ^E (J mol ^{−1})	μ (mPa s)	Δμ (mPa s)	ΔG ^E (J mol ^{−1})
T = 293.15 K						
0.0000	11.780	0.000	0.000	5.872	0.000	0.000
0.1016	9.561	−1.366	−104.160	4.788	−0.714	−116.505
0.2001	9.358	−0.742	19.242	4.937	−0.205	35.422
0.2991	9.146	−0.123	192.094	5.121	0.340	234.308
0.4008	9.790	1.375	472.632	5.250	0.839	437.845
0.5000	9.021	1.439	536.675	4.848	0.799	474.959
0.5992	8.127	1.378	549.871	4.225	0.538	426.159
0.7069	6.619	0.774	441.284	3.245	−0.050	219.136
0.7999	5.069	0.005	244.046	2.716	−0.240	89.521
0.8998	3.706	−0.519	−2.085	2.440	−0.151	33.645
1.0000	3.384	0.000	0.000	2.226	0.000	0.000
T = 303.15 K						
0.0000	8.079	0.000	0.000	4.436	0.000	0.000
0.1016	6.542	−0.976	−112.224	3.776	−0.402	−71.973
0.2001	6.559	−0.415	31.984	3.959	0.031	79.788
0.2991	6.527	0.100	208.462	3.902	0.226	212.221
0.4008	6.897	1.031	462.054	4.072	0.654	416.002
0.5000	6.516	1.199	543.084	3.743	0.578	438.124
0.5992	5.502	0.733	479.499	3.111	0.198	325.463
0.7069	4.468	0.293	351.820	2.328	−0.312	78.294
0.7999	3.696	0.035	227.635	1.961	−0.443	−58.234
0.8998	2.956	−0.353	−6.070	1.936	−0.214	−28.047
1.0000	2.756	0.000	0.000	1.895	0.000	0.000
T = 313.15 K						
0.0000	8.079	0.000	0.000	4.436	0.000	0.000
0.1016	6.542	−0.976	−112.224	3.776	−0.402	−71.973
0.2001	6.559	−0.415	31.984	3.959	0.031	79.788
0.2991	6.527	0.100	208.462	3.902	0.226	212.221
0.4008	6.897	1.031	462.054	4.072	0.654	416.002
0.5000	6.516	1.199	543.084	3.743	0.578	438.124
0.5992	5.502	0.733	479.499	3.111	0.198	325.463
0.7069	4.468	0.293	351.820	2.328	−0.312	78.294
0.7999	3.696	0.035	227.635	1.961	−0.443	−58.234
0.8998	2.956	−0.353	−6.070	1.936	−0.214	−28.047
1.0000	2.756	0.000	0.000	1.895	0.000	0.000
T = 323.15 K						
0.0000	8.079	0.000	0.000	4.436	0.000	0.000
0.1016	6.542	−0.976	−112.224	3.776	−0.402	−71.973
0.2001	6.559	−0.415	31.984	3.959	0.031	79.788
0.2991	6.527	0.100	208.462	3.902	0.226	212.221
0.4008	6.897	1.031	462.054	4.072	0.654	416.002
0.5000	6.516	1.199	543.084	3.743	0.578	438.124
0.5992	5.502	0.733	479.499	3.111	0.198	325.463
0.7069	4.468	0.293	351.820	2.328	−0.312	78.294
0.7999	3.696	0.035	227.635	1.961	−0.443	−58.234
0.8998	2.956	−0.353	−6.070	1.936	−0.214	−28.047
1.0000	2.756	0.000	0.000	1.895	0.000	0.000

of mixtures containing 1-butanol, *n*-hexadecane, and squalane.³³ The negative contribution (also known as random contribution) prevails when $0 < x_1 < 0.3$ and slightly when $0.9 < x_1 < 1$. It results from the disruption of the molecular order of the dilute component, resulting in the random separation of the molecules of the dilute component within the more concentrated component. On the other hand, the positive region (non-randomness region) ranges from values of the molar fraction of *n*-hexadecane of roughly 0.3–0.75. The positive values of the viscosity deviations are ascribed to van der

Waals molecular attractions and dispersive interactions occurring between permanent and induced dipoles. It is worthwhile noticing that as the temperature increases, the difference in the deviations between the positive and negative parts is less marked.³³

As for the variation of ΔG^E with the molar fraction, the values are mostly positive, with the exception of mixtures close to extreme composition. Positive values of this magnitude are indicative of the dominance of specific interactions among the molecules of the compounds in the binary mixture, whereas

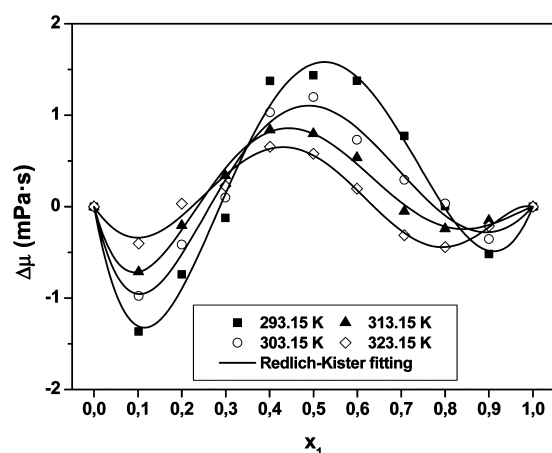


Figure 2. Viscosity deviations for the binary mixture of *n*-hexadecane (1) + solketal (2) at 293.15, 303.15, 313.15, and 323.15 K. Points show experimental observations, and the solid lines depict the Redlich–Kister fitting.

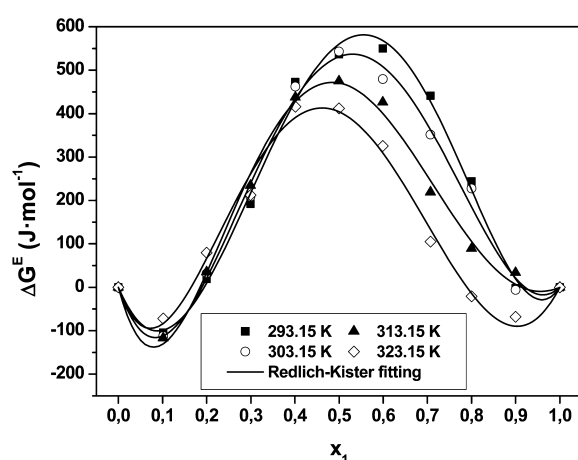


Figure 3. Deviation of the Gibbs free energy of activation of viscous flow for the binary mixture of *n*-hexadecane (1) + solketal (2) at 293.15, 303.15, 313.15, and 323.15 K. Points show experimental observations, whereas the solid lines represent the Redlich–Kister fitting.

negative values denote that dispersion forces prevail.⁵¹ Here, the effect of an increase of the temperature resulted in lower values of ΔG^\ddagger .

3.3. Tensiometric Properties. Surface tensions of the whole set of binary mixtures were determined at 298.15, 303.15, 313.15, and 323.15 K and $P = 0.1$ MPa. Table 5 lists the values obtained from the measurements as well as the surface tension deviations, whose plots can be seen in Figure 4 together with the corresponding Redlich–Kister correlated curve, whose coefficients are compiled in Table 3. The surface tension deviations $\Delta\gamma$ were calculated using eq 7

$$\Delta\gamma = \gamma - (x_1\gamma_1 + (1 - x_1)\gamma_2) \quad (7)$$

where γ is the surface tension of the mixture and γ_1 and γ_2 refer to those of cetane and solketal, respectively.

As seen in Figure 4, the $\Delta\gamma$ values are negative for all compositions of the binary mixtures, becoming less negative as the temperature was increased. It has been suggested that the excess surface tension is indicative of an uneven distribution of the components between the surface region and the bulk

Table 5. Surface Tension (γ) and Surface Tension Deviations ($\Delta\gamma$) for the Binary Mixtures of *n*-Hexadecane (1) + Solketal (2) at 298.15, 303.15, 313.15, and 323.15 K

x_1	γ (mN m ⁻¹)	$\Delta\gamma$ (mN m ⁻¹)	γ (mN m ⁻¹)	$\Delta\gamma$ (mN m ⁻¹)
$T = 298.15$ K				
0.0000	32.0788	0.0000	30.9144	0.0000
0.1016	26.8741	-4.7219	26.3195	-4.1180
0.2001	24.6068	-6.5211	23.9340	-6.0410
0.2991	23.6399	-7.0175	22.9933	-6.5170
0.4008	23.3890	-6.7854	22.7677	-6.2654
0.5000	23.5474	-6.1555	22.8942	-5.6731
0.5992	23.4418	-5.7895	22.8597	-5.2419
0.7069	23.5761	-5.1437	22.7760	-4.8203
0.7999	23.7268	-4.5510	23.0816	-4.0781
0.8998	24.0935	-3.7092	23.5778	-3.1125
1.0000	27.3268	0.0000	26.2202	0.0000
$T = 303.15$ K				
0.0000	31.5929	0.0000	30.2163	0.0000
0.1016	26.6503	-4.4780	25.8792	-3.8400
0.2001	24.3608	-6.3170	23.3251	-5.9120
0.2991	23.4204	-6.8047	22.3678	-6.3849
0.4008	23.2032	-6.5571	22.3592	-5.8961
0.5000	23.4147	-5.8918	22.3636	-5.4061
0.5992	23.3093	-5.5435	22.2648	-5.0194
0.7069	23.3813	-4.9792	22.2234	-4.5340
0.7999	23.5972	-4.3381	22.3777	-3.9248
0.8998	23.9263	-3.5518	22.9169	-2.8963
1.0000	27.0201	0.0000	25.3231	0.0000
$T = 313.15$ K				
0.0000	30.9144	0.0000	30.2163	0.0000
0.1016	26.3195	-4.1180	25.8792	-3.8400
0.2001	23.9340	-6.0410	23.3251	-5.9120
0.2991	22.9933	-6.5170	22.3678	-6.3849
0.4008	22.7677	-6.2654	22.3592	-5.8961
0.5000	22.8942	-5.6731	22.3636	-5.4061
0.5992	22.8597	-5.2419	22.2648	-5.0194
0.7069	22.7760	-4.8203	22.2234	-4.5340
0.7999	23.0816	-4.0781	22.3777	-3.9248
0.8998	23.5778	-3.1125	22.9169	-2.8963
1.0000	26.2202	0.0000	25.3231	0.0000
$T = 323.15$ K				
0.0000	30.2163	0.0000	30.2163	0.0000
0.1016	25.8792	-3.8400	25.8792	-3.8400
0.2001	23.3251	-5.9120	23.3251	-5.9120
0.2991	22.3678	-6.3849	22.3678	-6.3849
0.4008	22.3592	-5.8961	22.3592	-5.8961
0.5000	22.3636	-5.4061	22.3636	-5.4061
0.5992	22.2648	-5.0194	22.2648	-5.0194
0.7069	22.2234	-4.5340	22.2234	-4.5340
0.7999	22.3777	-3.9248	22.3777	-3.9248
0.8998	22.9169	-2.8963	22.9169	-2.8963
1.0000	25.3231	0.0000	25.3231	0.0000

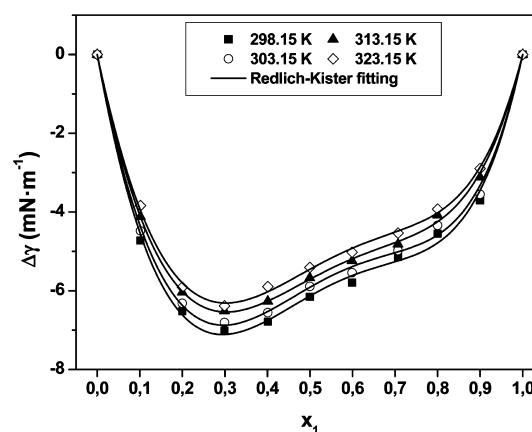


Figure 4. Surface tension deviations for the binary mixture of *n*-hexadecane (1) + solketal (2) at 293.15, 303.15, 313.15, and 323.15 K. Points show experimental observations, and solid lines represent the Redlich–Kister fitting.

region. Negative values of this magnitude represent that the components whose surface tensions are smaller have a higher concentration at the liquid surface than in the bulk in these mixtures.^{53,54} In this particular case, *n*-hexadecane has a lower surface tension than solketal, as indicated in Table 5, which translates into the fact that the former will tend to migrate to the surface, while solketal has a greater tendency to remain in the bulk phase.

3.4. Refractometric Properties. Finally, refractive indices have been measured at 293.15, 303.15, 313.15, and 323.15 K and $P = 0.1$ MPa. From the observed values of this property, the so-called Lorentz–Lorentz⁵⁵ molar refractivity R_m has been calculated, which is the most frequently used mixing rule in the

Table 6. Refractive Index (n_D), Molar Refractivity (R_m), and Lorentz–Lorentz Molar Refraction Index Deviations (ΔR_m) for the Binary Mixtures of *n*-Hexadecane (1) + Solketal (2) at 293.15, 303.15, 313.15, and 323.15 K

ϕ_1	n_D	R_m (m ³ mol ⁻¹)	ΔR_m (×10 ³ , m ³ mol ⁻¹)	ϕ_1	n_D	R_m (m ³ mol ⁻¹)	ΔR_m (×10 ³ , m ³ mol ⁻¹)
$T = 293.15$ K				$T = 313.15$ K			
0.0000	1.4345	0.0324	0.0000	0.0000	1.4305	0.0325	0.0000
0.0461	1.4345	0.0348	-2.1119	0.0466	1.4304	0.0354	-1.7018
0.1004	1.4342	0.0388	-2.4560	0.1017	1.4303	0.0392	-2.2820
0.1740	1.4344	0.0446	-1.0696	0.1750	1.4304	0.0451	-0.7624
0.2729	1.4344	0.0522	2.0452	0.2742	1.4303	0.0527	2.3450
0.3778	1.4340	0.0579	3.3160	0.3796	1.4299	0.0584	3.6684
0.4898	1.4344	0.0626	3.6999	0.4931	1.4301	0.0634	4.1883
0.6141	1.4332	0.0664	2.7158	0.6157	1.4298	0.0670	3.0617
0.7265	1.4342	0.0696	1.7692	0.7290	1.4291	0.0700	1.9556
0.8524	1.4346	0.0726	0.3850	0.8567	1.4286	0.0731	0.5559
1.0000	1.4347	0.0767	0.0000	1.0000	1.4300	0.0770	0.0000
$T = 303.15$ K				$T = 323.15$ K			
0.0000	1.4325	0.0324	0.0000	0.0000	1.4288	0.0327	0.0000
0.0464	1.4321	0.0349	-1.9722	0.0471	1.4286	0.0359	-1.3086
0.1014	1.4322	0.0388	-2.4484	0.1023	1.4286	0.0396	-2.0395
0.1749	1.4323	0.0448	-0.8546	0.1761	1.4286	0.0455	-0.4724
0.2735	1.4323	0.0523	2.1543	0.2744	1.4285	0.0529	2.4074
0.3788	1.4319	0.0580	3.4805	0.3807	1.4281	0.0589	3.8762
0.4920	1.4321	0.0629	3.9716	0.4941	1.4284	0.0638	4.3762
0.6139	1.4322	0.0665	2.8744	0.6175	1.4285	0.0676	3.3774
0.7269	1.4320	0.0696	1.8127	0.7299	1.4274	0.0704	2.0780
0.8543	1.4318	0.0727	0.4790	0.8568	1.4275	0.0735	0.6961
1.0000	1.4323	0.0766	0.0000	1.0000	1.4280	0.0773	0.0000

analysis of refractive index data.^{52,56} This variable can be calculated as follows:

$$R_m = V \frac{(n_D^2 - 1)}{(n_D^2 + 2)} \quad (8)$$

where V is the molar volume calculated from the density and molecular weights as indicated in eq 6 and n_D is the refractive index of the mixture.

Table 6 shows the refractive indices, the corresponding molar refractivities, and also the deviation of this property, ΔR_m , which is obtained as follows:

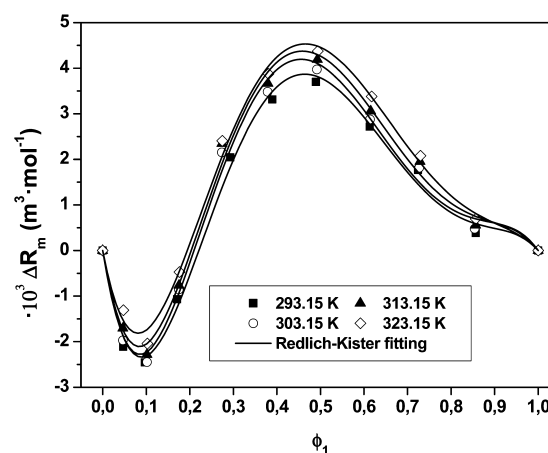
$$\Delta R_m = R_m - (\phi_1 R_1 + \phi_2 R_2) \quad (9)$$

where R_1 and R_2 are the molar refractivities of cetane and solketal, respectively, and ϕ_1 and ϕ_2 represent the volume fractions of the mentioned compounds, which may be calculated from the molar fractions, x_i , and molar volumes, V_i , with eq 10.

$$\phi_i = \frac{x_i V_i}{\sum_i x_i V_i} \quad (10)$$

Figure 5 represents the deviations derived from eq 9 with respect to the volume fraction of each of the components at each temperature tested, as customary for this property.^{51,56} As with the other properties, the Redlich–Kister fitted curve is shown, with the corresponding coefficients being present in Table 3.

The evolution of ΔR_m with changing compositions of the binary mixtures follow a S-shaped curve, although not too different from that observed in Figure 2 for the deviation of the viscosity. In this case, values are negative for compositions in the range of $0 < x_1 < 0.2$ and positive thereafter and the effect

**Figure 5.** Molar refractivity deviations for the binary mixture of *n*-hexadecane (1) + solketal (2) at 293.15, 303.15, 313.15, and 323.15 K. Points show experimental observations, and the line is the Redlich–Kister fitting.

of the temperature is slightly positive on the values of the deviation of the molar refractivity.

4. CONCLUSION

Comprehensive sets of experimental data have been obtained for binary mixtures of *n*-hexadecane as a model compound of diesel fuel and solketal, a novel bio-based fuel additive, which are of interest for potential equipment design. These experimental efforts include density and surface tension at 298.15, 303.15, 313.15, and 323.15 K as well as dynamic viscosity and refractive index at 293.15, 303.15, 313.15, and 323.15 K. From the experimental data, excess molar volume and the deviations of the properties were determined.

The excess volume and molar refractivity deviation showed a S-shaped curve behavior, while the viscosity deviation showed a W-shaped curve, thus acquiring negative and positive values at different composition values of the binary mixtures. The Gibbs free energy of activation for viscous flow, which is dependent upon molar volume and viscosity, also showed this type of behavior to some extent, although in a less marked way. Finally, the surface tension deviation proved negative at all compositions of the binary mixture.

Last, these deviations were fitted to a Redlich–Kister equation consisting of four parameters, thus allowing us to obtain the corresponding parameters and standard deviations of the calculated values with respect to the experimental observations.

AUTHOR INFORMATION

Corresponding Author

*Telephone: +44-0-121-414-5081. E-mail: j.estebanserrano@bham.ac.uk

Notes

The authors declare no competing financial interest.

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NOMENCLATURE

Chemicals

C₁₆ or 1 = *n*-hexadecane

Sk or 2 = solketal

Abbreviations

A_i = each of the parameters of the Redlich–Kister equation

G^E = excess Gibbs energy of activation for viscous flow

M = molecular weight

n = number of parameters used in the Redlich–Kister equation

N = number of experimental data

n_D = refractive index

P = excess or deviation property

R = ideal gas constant (J mol⁻¹ K⁻¹)

R_m = molar refractivity (m³ mol⁻¹)

T = temperature (K)

V = molar volume (m³ mol⁻¹)

V^E = molar excess volume (m³ mol⁻¹)

Greek Letters

γ = surface tension (mN m⁻¹)

Δ = relative to the deviations of each property

ϕ = volume fraction

μ = dynamic viscosity (mPa s)

ρ = density (g cm⁻³)

σ = standard deviation

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