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Mass Effect on the Soret Coefficient in *n*-alkane Mixtures

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We have determined the Soret coefficient of different equimolar and non equimolar *n*-alkane mixtures from measurements of the molecular diffusion and thermal diffusion coefficients. It is shown that equimolar mixtures behave as isotopic-like mixtures in which only the mass effect contributes to the Soret effect. In non equimolar mixtures, a small linear dependence with the molar fraction is observed. Finally, we have obtained a new correlation, which allows the determination of the Soret coefficient of *n*-alkane mixtures using the data of viscosity, the thermal expansion coefficient of the pure components and the density of the equimolar mixture.

I. INTRODUCTION

When a thermal gradient is applied to a homogeneous binary liquid mixture, a transport of matter is induced, which causes a concentration gradient. This phenomenon is known as thermal diffusion or the Soret effect. According to non equilibrium thermodynamics theory, when concentration and temperature gradients are present in a mixture of density ρ , the mass transport of one component is given as follows:

$$J = -\rho D \nabla c - \rho D_T c \ 1 - c \ \nabla T \tag{1}$$

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where c is the mass fraction of the reference component, and D and D_T are the mass diffusion coefficient and the thermal diffusion coefficient, respectively. In the non equilibrium stationary state, J=0 and then the resulting concentration gradient is

$$\nabla c = -\frac{D_T}{D}c \ 1 - c \ \nabla T \tag{2}$$

Thus, the amplitude of thermal diffusion separation is determined by the following ratio $S_T = (D_T/D)$, which is known as the Soret coefficient. In gaseous mixtures, the thermal diffusion factor $(\alpha_T = S_T T)$ is more commonly used.

Many efforts have been made to determine the molecular parameters responsible for the Soret effect. In low-density systems, the kinetic theory provides expressions for the thermal diffusion factor. Chapmann¹ showed that in isotopic mixtures of hard spheres, α_T could be expanded in powers of the relative mass difference between species. Kincaid *et al.*² found that such an expansion exists for all fluid densities. More recently, Galliero *et al.*³ have shown from molecular dynamics simulation for isotope-like mixtures of Lennard-Jones spheres that the Chapmann expansion holds at any mass ratio and density. On the other hand, Debuschewitz and Köhler⁴ and Wittko and Köhler⁵ have experimentally shown that in isotopic mixtures of different liquids the Soret coefficient can be split into different additive contributions. One contribution, the so-called isotopic effect, stems from the differences in both masses and moments of inertia between the molecules and is independent of composition. An additional chemical contribution depending on concentration accounts for the chemical difference between the two species. Recently, Hartman *et al.*^{6,7} were able to experimentally show that the isotope contribution is also present in mixtures of different liquids. Artola and Rousseau⁸ have found from dynamic simulations in a Lennard-Jones fluid that the chemical contributions is due to the cross interaction between unlike molecules. Finally, Leroyer and Würger^{9,10} derived an expression for the Soret coefficient, which accounts for several features observed in experiments.

We focus in this work on the Soret coefficient in n-alkane mixtures. Normal alkanes are almost ideal mixtures with rather simple physical properties which made them appealing for thermal diffusion. In previous works¹¹⁻¹⁴, we have studied the mass and the thermodiffusion coefficient obtaining empirical correlations. In the following, we shall show that the mass effect is the relevant contribution to the Soret coefficient in mixtures of n-alkanes.

II. EXPERIMENTAL SECTION

All the products used in this study were purchased from Merck with purity higher than 99%. The mixtures have been prepared by weight using a Gram VXI-310 digital scale with a precision of 0.001 g.

The relevant thermophysical properties of the studied mixtures were measured as follows: An Anton Paar DMA 5000 densimeter with an accuracy of 5×10^{-6} g/cm³ was used to determine the density (ρ). The thermal expansion coefficient, α =- $(1/\rho)(\delta\rho/\delta T)$, was obtained by measuring the density of a mixture at different temperatures near the working temperature. Finally, the dynamic viscosity μ was measured with an Anton Paar AMVn falling ball microviscometer with reproducibility better than 0.1%. Some of these properties used in this work were taken from previous studies^{11,15,16}, while the rest where measured for this work. The values of the thermophysical properties are shown in Table I (equimolar mixtures) and Table II (non equimolar mixtures). All measurements were done at 298 K.

The thermal diffusion coefficient was determined using the thermogravitational column (TGC) technique ^{11,17}. The working principles of a TGC are as follows: The combination of horizontal separation by thermal diffusion with vertical convection currents produces a separation of the components between the column ends. The theory for TGC¹⁸ establishes a relationship between the stationary separation and the thermal diffusion coefficient D_T . In the following we shall take as the reference component the heavier one that will be denoted by subindex 1. In n-alkane mixtures, D_T >0 for this component. Then, this relation can be written as follows:

$$\Delta c = \frac{504 L_z}{g L_x^4} \frac{D_T v}{\alpha} c_0 \ 1 - c_0 \tag{3}$$

where Δc is the mass fraction difference between the bottom and the top of the column, L_z is the height of the TGC, L_x is the gap of the TGC, c_0 is the mass fraction of the reference component in the initial homogeneous binary mixture, α the thermal expansion coefficient, ν is the kinematic viscosity and g is the acceleration due to gravity. For a summarized explanation see Refs. 11 and 17.

The TGC used in this work was a stainless steel concentric tube column closed at both ends. The geometrical parameters of the column were the height L_z =0.5±0.001 m and a gap L_x =1× 10⁻³ ± 5×10⁻⁶ m. The temperature difference across the column was 10 K with an average temperature of 298 K.

To experimentally determine the mass separation between the column ends, a preliminary calibration is carried out, that relates the mass fraction with a physical property of the mixture: density in the present case. The calibration is made, measuring the density of several mixtures of known composition near the one of the standard mixture. A

linear relation between the density and the mass fraction is obtained in all the cases. Thus, the mass fraction with a deviation around ± 0.01 wt% is determined.

For the determination of the molecular diffusion coefficient, the sliding symmetric tubes (SST) technique, which has been validated with the experimental data of several binary mixtures, was used ^{14,19}. This method consists of several sets of two identical vertical tubes, each containing a mixture with a slightly different mass concentration. The denser mixture is placed in the lower tube in order to eliminate convection. At the beginning of the experiment, the sets in their separated configuration are placed in a water bath until they all reach the working temperature. The sets are then changed to the faced configuration, allowing the diffusion to start. At different times, they are changed one by one back to the separated configuration to stop the mass transfer. The densities of the mixtures are measured, and the average concentration inside each tube is determined from a calibration curve. Thus, the change in average concentration with time is obtained for both tubes and, consequently, the molecular diffusion coefficient can be calculated from the solution of the equations of approach to equilibrium. The procedure has been described in detail in earlier publications ^{14,19}.

III. RESULTS AND DISCUSSION

A. Soret coefficient for equimolar mixtures

In this work 29 *n*-alkane equimolar mixtures corresponding to the following series have been considered:

- nC_6 - nC_i with i=10, 11, 12, 13, 14, 15, 16, 17, 18;
- nC_{10} - nC_i with i=5, 7, 14, 15, 16, 17, 18;
- nC_{12} - nC_i with i=5, 7, 8, 9, 16, 17, 18;
- nC_{18} - nC_i with i=5, 7, 8, 9, 11, 13.

Table I summarizes the values of the measured mass diffusion coefficients at 298 K for the equimolar mixtures considered. Table I also shows the values of the thermal diffusion coefficient for these mixtures. The values for the systems nC_6 - nC_{11} , nC_6 - nC_{12} , nC_6 - nC_{13} , nC_6 - nC_{15} , nC_6 - nC_{17} , nC_{10} - nC_{14} and nC_{12} - nC_{16} have been measured in this work. D_T values for the other systems have been taken from Ref. 11. With the values of D and D_T , the Soret coefficient (S_T = D_T /D) of the mixtures has been determined, which is also detailed in Table I.

The kinetic theory provides expressions that adequately describe the behaviour of the thermal diffusion factor in the case of low-density systems. Thus, it is found that in isotopic mixtures of hard spheres α_T exhibits a relative simple dependence on (M_I/M_2) , where M_I and M_2 are the masses of components of the mixture. Chapman¹ showed that for equimolar mixtures and in the Boltzman limit it is possible to expand α_T in powers of relative mass difference, $\delta M = (M_1 - M_2)/(M_1 + M_2)$.

$$\alpha_T = \alpha_0 \delta M + \cdots \tag{4}$$

Later Kinkaid et al. ² found that this expansion is valid at all fluid densities.

More recently, Galliero *et al.*³ analysed the validity of Eq. (4) for the thermal diffusion factor using molecular dynamics simulations for isotopic-like mixtures of Lennard-Jones spheres, which differ only in the mass between species. They found that this expression holds for all the studied conditions, with the coefficient α_0 depending strongly on density and weakly on temperature.

Even if n-alkanes are long and flexible molecules far from being spherical, it has been shown that the thermal diffusion results using Lennard-Jones spheres present a similar trend to those obtained with more realistic models. Hence in what follows we shall take the relative mass difference and the density as the relevant parameters to analyse the experimental results of the Soret coefficient. On the other hand as it has been pointed out by Polyakov et $al.^{20}$, in n-alkanes it is difficult to separate the contribution of the mass and moment of inertia because the moment of inertia increases almost linearly with the chain length (i.e. with the molecular mass).

Assuming, as suggested in Ref. 3, a fourth-power density dependence for α_0 , we have plotted in Fig.1 the obtained values of the Soret coefficient for equimolar mixtures versus the product $\rho^4 \delta M$. As can be seen, the data fits well to a quadratic curve given by

$$S_T = 0.0464 \ \rho^4 \delta M \quad 1 - 1.57 \ \rho^4 \delta M \tag{5}$$

where the density is expressed in g/cm³. The second factor in this equation amounts to a correction of a few percent for the mixtures considered.

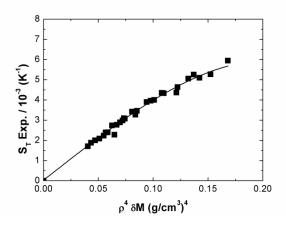


FIG. 1. Experimental Soret coefficient in function of the $\rho^4 \delta M$ factor, for binary equimolar mixtures

According to Eq. (5), S_T increases with density and relative mass difference. The difference between the S_T values calculated from Eq. 5 and the experimental ones are in all the cases small, and lower than the experimental error. Therefore Eq. 5 permits the quantitative determination of the Soret coefficient of an equimolar mixture of n-alkanes from the density measurements of the mixture.

Taking typical values for densities of the considered mixtures of ρ =0.70 g/cm³, the values obtained from Eq. (5) for α_0 is α_0 =3.3 (α_0 =0.0464 ρ^4 T), which is of the same order as the one calculated by Galliero et al³ in the liquid-state region.

From the above results, we can conclude that equimolar *n*-alkane mixtures behave as isotopic-like mixtures with the mass effect being the sole contribution to the Soret coefficient.

B. Dependence of the Soret coefficient on concentration

In addition, this work has been completed by studying the systems nC_{12} - nC_6 , nC_{12} - nC_7 , nC_{12} - nC_8 and nC_{10} - nC_5 at the following concentrations:

- nC_{12} - nC_6 (x_1 =0.10, 0.30, 0.34, 0.50, 0.70, 0.90);
- nC_{12} - nC_7 (x_1 =0.10, 0.30, 0.37, 0.50, 0.70, 0.90);
- nC_{12} - nC_8 (x_1 =0.07, 0.31, 0.40, 0.50, 0.73);
- nC_{10} - nC_5 (x_1 =0.10, 0.20, 0.34, 0.50, 0.64, 0.80).

where x_1 is the molar fraction of the heavier component.

Table II shows the measured values of D and D_T at 298 K and different concentrations for the four systems considered. The mass diffusion coefficient for the system nC_{10} - nC_5 and some of the thermal diffusion coefficients for nC_{12} - nC_6 and nC_{12} - nC_8 were measured in this work. The remaining values of D and D_T were taken from Refs. 11-13, 15 and 16. From the values of D and D_T , we determined the Soret coefficient of the different mixtures. We have plotted in Fig. 2 these values of the Soret coefficient at different composition for the four systems.

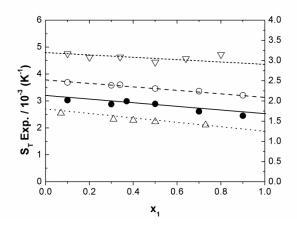


FIG. 2. Soret coefficient in function of the molar fraction of the heavier component, x_I , for the systems "o" nC_{12} - nC_6 , "•" nC_{12} - nC_7 and " Δ " nC_{12} - nC_8 and " ∇ " nC_{10} - nC_5 at 298 K. The solid straight lines indicate values for the Soret coefficient calculated from Eq. 17 as described in the text, the symbols represent experimental results from this work. The label on the right axis corresponds to the mixture nC_{10} - nC_5 .

In previous works^{12,14}, we have shown that both the molecular diffusion and the thermal diffusion coefficients vary linearly with the mass fraction in n-alkane mixtures, being this variation strong. However, in the following we shall show that the Soret coefficient presents a small linear dependence with the molar fraction. This behaviour has also been seen in other works²¹.

According to Refs. 12 and 14, the coefficients D and D_T can be written as follows:

$$D = D_1^0 c_2 + D_2^0 c_1; \ D_T = D_{T1}^0 c_2 + D_{T2}^0 c_1$$
 (6)

where D_1^0 , D_2^0 , D_{TI}^0 and D_{T2}^0 are the corresponding infinite dilution values, which are defined by

$$D_i^0 = \lim_{c_i \to 0} D; \ D_{Ti}^0 = \lim_{c_i \to 0} D_T \tag{7}$$

Accordingly, the Soret coefficient, $S_T = D_T/D$, expressed in terms of molar fractions x_1 and x_2 is:

$$S_T = \frac{D_{11}^0 M_2 x_2 + D_{12}^0 M_1 x_1}{D_{11}^0 M_2 x_2 + D_{21}^0 M_1 x_1} \tag{8}$$

In Ref. 14, we have shown that the ratio D_1^0/D_2^0 is equal to the ratio of molecular masses, M_1/M_2 . Thus,

$$D_1^0 M_2 = D_2^0 M_1 \tag{9}$$

Therefore, Eq. (11) can be written as

$$S_T = S_{T1}^0 x_2 + S_{T2}^0 x_1 \tag{10}$$

where $S_{T1}^{\ 0}$ and $S_{T2}^{\ 0}$ are the limiting Soret coefficients given by

$$S_{Ti} = \frac{D_{Ti}^0}{D_i^0} \tag{11}$$

Eq. (10) shows that the Soret coefficient in n-alkane mixtures is indeed a linear function of the molar fraction. In Fig. 2, the Soret coefficient for the nC_{12} - nC_6 , nC_{12} - nC_7 , nC_{12} - nC_8 and nC_{10} - nC_5 systems is represented as a function of the molar fraction of the heavier component. As one can see, the Soret coefficient for each system fits well to a straight line in accordance with Eq. (10). A linear dependence also occurs in isotopic mixtures of hard spheres².

Eq. (10) can be written more conveniently in terms of the Soret coefficient of the equimolar mixture that we shall denote by (S_T) and the difference $\Delta S_T = (S_{T1}^0 - S_{T2}^0)/2$. Then we have,

$$S_T = S_T \ 1 - \lambda \ x_1 - x_2 \tag{12}$$

The coefficient λ is

$$\lambda = \frac{\Delta S_T}{S_T} = \frac{S_{T_1}^0 - S_{T_2}^0}{S_{T_1}^0 + S_{T_2}^0} \tag{13}$$

which, taking into account Eqs. (9) and (11), can be written as

$$\lambda = \frac{D_{T_1}^0 M_2 - D_{T_2}^0 M_1}{D_{T_1}^0 M_2 + D_{T_2}^0 M_1} \tag{14}$$

where $D_{T1}^{\ 0}$ and $D_{T2}^{\ 0}$ can be calculated from the expressions obtained in Ref. 12:

$$D_{T1}^{0} = 5.34 \cdot 10^{-11} \frac{\alpha_2}{\mu_2} M_2 1 - \frac{M_2}{M_1}$$
 (15)

$$D_{T2}^{0} = 3.71 \cdot 10^{-11} \frac{\alpha_{1}}{\mu_{1}} M_{1} \frac{M_{1}}{M_{2}} - 1$$
 (16)

where α_i and μ_i are the thermal expansion coefficient and the dynamic viscosity of the pure components, respectively. The subindexes 1 and 2 refer to the heavier and lighter components of the mixture, respectively. Table III displays the values obtained for D_{T1}^{0} , D_{T2}^{0} and λ using Eqs. (14), (15) and (16) for the binary systems considered. The values of the Soret coefficient calculated with Eq. (17) for the different concentrations appear in Table II. As can be seen, the agreement with the corresponding experimental values is excellent. Therefore, the dependence of S_T on concentration is determined by the thermophysical properties α and μ of the pure components and their molecular mass. Eq. (17) shows that this dependence is governed by the factor λ , which is a positive number smaller than unity on the order of 10^{-1} . Therefore, S_T decreases with the increasing molar fraction of the heavier component. Furthermore, large values of λ mean a strong dependence of S_T on concentration. For example, for the system nC_{10} - nC_5 λ =0.040, indicating that S_T is practically independent of composition. However, for the nC_{12} - nC_8 system λ takes the value 0.171, pointing out a higher dependence of S_T on the molar fraction. These results are corroborated by our experimental results (see Fig. 2).

In any case, λ being small, S_T changes little with composition. According to Hartmann *et al.*⁷, this means that the so-called chemical contribution to S_T is small corresponding to the chemical similarity of the molecules. The main contribution to S_T comes, in *n*-alkane mixtures, from the mass differences between the molecules.

IV. CONCLUSIONS

In the first part of this work, we determined the Soret coefficient of 29 equimolar mixtures of n-alkanes from measurements of thermal diffusion and molecular diffusion coefficients. We showed that a quadratic relation between the Soret coefficient and the relative mass difference exists, with the coefficient depending strongly on density. Thus, equimolar n-alkane mixtures behave as isotopic-like mixtures in which only the mass effect contributes to the Soret coefficient.

In the second part, we determined the Soret coefficient of four binary systems in the whole range of concentrations. It was showed that the Soret coefficient presents a small linear dependence on the molar fraction, decreasing with the concentration of the heavier component. A correlation is derived that allows the quantitative determination of the Soret coefficient of a mixture of n-alkanes at any concentration from the viscosity, the thermal expansion coefficients of pure components and the density of the equimolar mixture.

V. ACKNOWLEDGMENTS

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REFERENCES

- ¹ S. Chapman, Proc. R. Soc. Lond. **177**, 38 (1940).
- ² J.M. Kincaid, E.G.D. Cohen and M. Lopez de Haro, J. Chem. Phys. **86**, 963 (1987).
- ³ G. Galliero, M. Bugel, B. Duguay and F. Montel, J. Non-Equilib. Thermodyn. **32**, 251 (2007).
- ⁴ C. Debuschewitz and W. Köhler, Phy. Rev. Lett. **87**, 055901-1 (2001).
- ⁵ G. Wittko and W. Köhler, J. Chem. Phys. **123**, 14506 (2005).
- ⁶ S. Hartmann, G. Wittko, W. Köhler, K.I. Morozov, K. Albers and G. Sasowski, Phys. Rev. Lett. **109**, 065901 (2012).
 - ⁷ S. Hartmann, W. Köhler and K.I. Morozov, Soft Matter **8**, 1355 (2012).
 - ⁸ P. Artola and B. Rousseau, Phys. Rev. Lett. **98**, 125901 (2007).
 - ⁹ Y. Leroyer and A. Würger, J. Chem. Phys. **135**, 054102 (2011).
 - ¹⁰ A. Würger, Phys. Rev. Lett. **102**, 078302 (2009).
- ¹¹ P. Blanco, M.M. Bou-Ali, J.K. Platten, P. Urteaga, J.A. Madariaga and C. Santamaría, J. Chem. Phys. **129**, 174504 (2008).
- ¹² J.A. Madariaga, C. Santamaría, M.M. Bou-Ali, P. Urteaga and D. Alonso de Mezquia, J. Phys. Chem. B **114**, 6937 (2010).
 - ¹³ P. Blanco, P. Polyakov, M.M. Bou-Ali and S. Wiegand, J. Phys. Chem. B **112**, 8340 (2008).
- ¹⁴ D. Alonso de Mezquia, M.M. Bou-Ali, M. Larrañaga, J.A. Madariaga and C. Santamaría, J. Phys. Chem. B **116**, 2814 (2012).
 - ¹⁵ S. Srinivasan, D. Alonso de Mezquia, M.M. Bou-Ali and M.Z. Saghir, Philos. Mag. **91**, 4332 (2011).
 - ¹⁶ Y. Yan, P. Blanco, M.Z. Saghir and M.M. Bou-Ali, J. Chem. Phys. **129**, 194507 (2008).

¹⁷ J. Valencia, M. Bou-Ali, O. Ecenarro, J. Madariaga and C. Santamaria in Thermal Nonequilibrium Phenomena in Fluid Mixtures edited by W. Köhler and S. Wiegand (Springer, Berlin, 2002), p. 233.

¹⁸ W.H. Furry, R.C. Jones and L. Onsager, Phys. Rev. E **55**, 1083 (1939).

¹⁹ A. Mialdun, V. Yansou, V. Shevtsova, A. Königer, W. Köhler, D. Alonso de Mezquia and M.M. Bou-Ali, J. Chem. Phys. **135**, 244512 (2012).

²⁰ P. Polyakov, J. Luettermer-Strathmann and S. Wiegand, J. Phys. Chem. B **110**, 26215 (2006).

²¹ A. Würger. J. Phys. Condens. Matter. **26**, 035105 (2013)

TABLE I. Experimental values of ρ , α , μ , D, D_T and S_T at 298 K for equimolar mixtures. δM is the relative mass difference, S_T (cc) stands for S_T calculated from Eq. (5) and Δ is the relative difference between S_T and S_T (cc).

	0		α / 10 ⁻³	$\mu / 10^{-3}$	$D_T/10^{-12}$	$D/10^{-9}$	$S_T / 10^{-3}$	$S_T(cc) / 10^{-3}$	
Mixture	ρ (g/cm ³)	δM	(\mathbf{K}^{-l})	$(Pa \cdot s)$	(m^2/sK)	(m^2/s)	(K^{-1})	(K^{-1})	$\Delta(\%)$
	(5/6111)		(11)	, ,	1CC				
C	0.600500	0.24556	1 150	0.538	$1: nC_6 - nC_i$ 6.08^a	2.52	2.40	2.47	20
nC_{10} - nC_6	0.698508	0.24556	1.159			2.53	2.40	2.47	-2.8
$nC_{11}-nC_6$	0.706712	0.28918	1.124	0.623	6.40	2.14	2.99	2.97	0.7
$nC_{12}-nC_6$	0.713777	0.32808	1.094	0.711	6.69	1.93	3.46	3.43	0.9
$nC_{13}-nC_{6}$	0.720307	0.36290	1.068	0.831	6.61	1.67	3.96	3.84	3.1
$nC_{14}-nC_{6}$	0.726124	0.39431	1.047	0.945	6.68 ^a	1.54	4.34	4.21	3.1
nC_{15} - nC_{6}	0.731762	0.42277	1.025	1.108	6.47	1.48	4.37	4.56	-4.2
nC_{16} - nC_{6}	0.736556	0.44868	1.009	1.260	6.38 ^a	1.26	5.06	4.86	4.1
nC_{17} - nC_6	0.741007	0.47200	0.991	1.432	6.32	1.24	5.10	5.13	-0.6
nC_{18} - nC_6	0.745086	0.49407	0.976	1.715	5.90 ^a	1.12	5.27	5.38	-2.0
					2: nC_{10} - nC_{i}				
nC_{10} - nC_5	0.689961	0.32705	1.203	0.486	8.78 ^a	2.86	3.07	3.04	1.0
nC_{10} - nC_7	0.706677	0.17354	1.122	0.608	3.90^{a}	2.07	1.88	1.87	0.5
nC_{14} - nC_{10}	0.745410	0.16470	0.976	1.410	1.84	0.88	2.09	2.17	-3.7
nC_{15} - nC_{10}	0.749433	0.19774	0.960	1.582	2.15^{a}	0.78	2.74	2.61	4.7
$nC_{16}-nC_{10}$	0.752850	0.22827	0.952	1.762	2.23^{a}	0.72	3.10	3.01	3.0
$nC_{17}-nC_{10}$	0.756111	0.25652	0.939	2.009	2.29^{a}	0.70	3.27	3.38	-3.3
nC_{18} - nC_{10}	0.759213	0.28279	0.928	2.255	2.38^{a}	0.61	3.90	3.77	3.4
				Set	3: nC_{12} - nC_{i}				
nC_{12} - nC_5	0.706503	0.40492	1.126	0.671	8.81^{a}	2.20	4.00	3.94	1.5
nC_{12} - nC_{7}	0.720338	0.25926	1.066	0.807	4.74^{a}	1.64	2.89	2.89	0.0
nC_{12} - nC_{8}	0.726279	0.19717	1.043	0.892	3.23^{a}	1.45	2.23	2.33	-4.3
nC_{12} - nC_{9}	0.731708	0.14096	1.021	0.992	2.15^{a}	1.26	1.71	1.76	-2.8
nC_{16} - nC_{12}	0.759300	0.14141	0.930	2.113	1.16	0.58	2.00	2.02	-0.1
$nC_{17}-nC_{12}$	0.759557	0.17029	0.922	2.389	1.29^{a}	0.54	2.39	2.40	-0.4
$nC_{18}-nC_{12}$		0.19813	0.909	2.665	1.33^{a}	0.48	2.77	2.77	0.0
10 12				Set	4: <i>n</i> C ₁₈ - <i>n</i> C _i				
nC_{18} - nC_{5}	0.740784	0.55824	0.991	1.592	7.38 ^a	1.24	5.95	5.74	3.7
$nC_{18}-nC_{7}$	0.749065	0.43502	0.963	1.735	$5.00^{\rm a}$	0.95	5.26	4.99	5.4
$nC_{18}-nC_{8}$	0.752698	0.38053	0.948	1.875	3.94 ^a	0.85	4.64	4.58	1.3
$nC_{18}-nC_{9}$	0.756092	0.32985	0.939	2.045	3.00 ^a	0.69	4.35	4.16	4.6
$nC_{18} nC_{11}$	0.762256	0.23902	0.920	2.445	1.78 ^a	0.52	3.42	3.27	4.6
$nC_{18} nC_{11}$ $nC_{18} nC_{13}$		0.15982	0.906	2.945	0.98^{a}	0.43	2.28	2.35	-3.0
Values of I		0.13702	0.700	2.7 13	0.70	0.15	2.20	2.33	5.0

^a Values of D_T taken from Ref. 11.

TABLE II. Experimental values of D, D_T and S_T at 298 K for non equimolar mixtures. S_T (cc) stands for S_T calculated with Eq. (17) and Δ is the relative difference between S_T and S_T (cc).

3.6		ρ	$\alpha / 10^{-3}$	$\mu / 10^{-3}$	$D_T/10^{-12}$	$D / 10^{-9}$	$S_T / 10^{-3}$	$S_T(cc) / 10^{-3}$	1 (0()
Mixture	x_1	(g/cm^3)	(K^{-1})	(Pa·s)	(m^2/sK)	(m^2/s)	(K^{-1})	(K^{-1})	Δ(%)
nC_{12} - nC_6	0.10	0.671330	1.240	0.360	9.14	2.48	3.69	3.70	-0.3
	0.30	0.695112	1.174	0.524	8.02	2.24	3.58	3.56	0.5
	0.34	0.698780	1.158	0.563	7.67	2.13	3.60	3.54	1.6
	0.50	0.713777	1.094	0.711	6.69	1.93	3.46	3.43	0.9
	0.70	0.728268	1.035	0.940	5.79	1.72	3.36	3.21	4.6
	0.90	0.740650	0.990	1.255	4.85	1.51	3.21	3.15	1.9
nC_{12} - nC_7	0.10	0.689767	1.202	0.475	6.34^{a}	2.09	3.03	3.22	-5.9
	0.30	0.706600	1.124	0.611	5.30^{a}	1.84	2.88	3.05	-5.6
	0.37	0.711000	1.102	0.682	5.08^{a}	1.70	2.99	2.99	0.0
	0.50	0.720338	1.066	0.807	4.74°	1.64	2.89	2.89	0.0
	0.70	0.731471	1.023	1.004	3.86^{a}	1.48	2.61	2.72	-4.0
	0.90	0.740949	0.990	1.234	3.35^{a}	1.37	2.45	2.55	-3.9
nC_{12} - nC_8	0.07	0.703400	1.146	0.555	4.34	1.70	2.55	2.67	-4.5
	0.31	0.717320	1.101	0.734	3.62	1.56	2.32	2.48	-6.5
	0.40	0.721960	1.060	0.807	3.39^{d}	1.49	2.28	2.41	-5.4
	0.50	0.726279	1.043	0.892	3.23^{c}	1.45	2.23	2.33	-4.3
	0.73	0.735870	1.022	1.105	2.64	1.25	2.11	2.15	1.9
nC_{10} - nC_5	0.10	0.637718	1.495	0.277	11.90^{b}	3.75	3.17	3.14	1.0
	0.20	0.652835	1.399	0.333	10.36 ^b	3.36	3.08	3.12	-1.3
	0.34	0.670325	1.297	0.403	$9.37^{\rm b}$	3.03	3.09	3.08	0.3
	0.50	0.689961	1.203	0.486	$8.76^{\rm c}$	2.96	2.96	3.04	-2.6
	0.64	0.700261	1.150	0.586	7.56^{b}	2.48	3.05	3.01	1.3
a v / - 1	0.80	0.711864	1.098	0.690	6.92 ^b	2.20	3.15	2.97	6.0

^a Values of D_T taken from Ref. 12; ^b Ref. 13; ^c Ref. 11; ^d Ref. 16. Values for D coefficient have been taken from Ref. 14 with the exception of the values of nC_{10} - nC_5 system, measured in this work.

TABLE III. Values of the limiting values of D_{TI}^0 , D_{T2}^0 and λ of nC_{10} - nC_5 , nC_{12} - nC_6 , nC_{12} - nC_7 and nC_{12} - nC_8 mixtures.

Mixture	$D_{TI}^{0}/10^{-12}$ (m ² /sK)	$D_{T2}^{0} / 10^{-12}$ (m ² /sK)	λ
nC_{10} - nC_5	13.65	6.37	0.041
nC_{12} - nC_6	10.46	4.34	0.100
nC_{12} - nC_7	7.06	3.11	0.144
nC_{12} - nC_{8}	4.56	2.18	0.171