

Effect of Thermophysical Properties and Morphology of the Molecules on Thermodiffusion Coefficient of Binary Mixtures

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Received: 18 December 2013 / Accepted: 5 May 2014 / Published online: 27 May 2014
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Abstract In this article, new experimental data of the thermal diffusion coefficient (D_T) of 20 binary mixtures of hexane-hexadecane, decane-hexadecane, toluene-hexadecane and 1-metilnaphthalene-hexadecane at several different compositions and at 298K and atmospheric pressure, are reported. Thermal diffusion coefficients were measured in a thermogravitational column with rectangular configuration. The results obtained show that the mass fraction dependence of thermodiffusion coefficients of the mixture is linear in all the cases. The studied mixtures have a common component, hexadecane, and they can be classified into two groups according to their mass and to the morphology of their components. We also show that the thermal diffusion coefficient and mixture viscosity are related in a different way for mixtures of n-alkanes and for mixtures of aromatic rings.

Keywords Thermophysical properties · Thermodiffusion coefficient · Thermogravitational technique · Binary mixtures

Introduction

The phenomenon of thermodiffusion has generated a great interest in the scientific community, due to its appearance

in numerous processes both natural and industrial of different fields such as petroleum industry (Capuano et al. 2011; Tello Alonso et al. 2012; Montel 1998).

In the case of binary mixtures, there are a lot of techniques which allow the determination of the thermodiffusion coefficient, such as Thermal Diffusion Forced Rayleigh Scattering (TDFRS) (Leppla and Wiegand 2003), Optical Beam Deflection (OBD) (Königer et al. 2009), Optical Digital Interferometry (ODI) (Mialdun and Shevtsova 2011), modern light scattering techniques (Crocco et al. 2012) or the Thermogravitational technique (TGC) (Bou-Ali et al. 2003). These experimental techniques were compared and validated by the performance of a Benchmark for binary mixtures (Platten et al. 2003).

The analysis of this phenomenon becomes considerably more complicated when mixtures of more than two components are studied. In that case, orbital laboratories are an ideal environment for the performance of the experiments thanks to the absence of the convection produced by the gravity.

Due to this increasing interest, a cooperative international project supported by the European Space Agency (ESA) was developed. In it, scientists expect to obtain reliable benchmark results by the SODI instrument in the International Space Station (ISS) and validate their ground-based techniques (Mialdun et al. 2012).

With the aim of clarifying the behaviour of the thermodiffusion phenomenon in multicomponent mixtures, studies in binary mixtures about the influence of different properties in the thermodiffusion phenomenon have been carried out. In Madariaga et al. (2010) a correlation that allows the determination of the thermodiffusion coefficient in binary mixtures of n-alkanes at any concentration from the dynamic viscosity, the thermal expansion coefficient and the molecular weight of the mixture was developed. In Leahy-Dios

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and Firoozabadi (2007) the influence of factors such as the size and the shape of the molecules was analyzed, and later, in Leahy-Dios et al. (2008) the influence of the viscosity was analyzed too. In addition, in other works, the influence of the moment of inertia and the chemical effects was also studied experimentally, by modelling or by simulations (Debuschewitz and Köhler 2001; Villain-Guillot and Würger 2011; Galliéro et al. 2003).

The motivation of this work is to try to expand the knowledge about the influence of different properties on the thermodiffusion coefficient. Therefore, the objective of this work is to analyze the influence of the molecular weight, the viscosity and the morphology of the molecules on the thermodiffusion coefficient.

With the aim of achieving this objective, 4 binary mixtures have been chosen with a common component: hexadecane.

This work is divided as follows. In Section “[Experimental Methodology](#)”, the experimental methodology followed is described; in Section “[Results and Discussion](#)” the obtained results and the discussion about them are shown. Finally, in Section “[Conclusions](#)” the conclusions achieved are shown.

Experimental Methodology

Studied Mixtures

In this work the thermogravitational behaviour of four binary systems with different concentrations has been studied. The four systems have a common component, hexadecane. The analyzed systems are: hexadecane-hexane, hexadecane-decane, hexadecane-toluene and hexadecane-1-methylnaphtalene. From here on, hexadecane will be referred as nC16, hexane as nC6, decane as nC10, toluene as Tol and 1-methylnaphtalene as MN. These systems can be grouped by their molecular weight (on the one hand nC16-nC6 and nC16-Tol and on the other hand nC16-nC10 and nC16-MN) and by the morphology of their components (nC16-nC6 and nC16-nC10 are chains and nC16-Tol and nC16-MN are a chain and an aromatic ring). Each system was studied at different concentrations of nC16.

Table 1 shows the concentrations of nC16, the difference of molecular weight between the components of the mixture, $\Delta M = M_{nC16} - M_i$, where M_i is the molecular weight of the second component (Tol, MN, nC6 or nC10), and the configuration of the mixture.

Every product used in this work was purchased from Merck and Aldrich, with purity higher than 99 %. The concentrations of the binary mixture were obtained by weight in a scale with accuracy of 0.0001 g. The necessary volume of mixture for each experiment was of approximately 30

Table 1 Description of the analyzed concentrations, the difference of molecular weight and configuration of each mixture

nC16-i	<i>c</i> (nC16)	ΔM	Config,
nC6	0.20	140.27	Chain-Chain
	0.40		
	0.50		
	0.72		
Tol	0.20	134.31	Chain-Ring
	0.30		
	0.40		
	0.50		
	0.60		
	0.80		
nC10	0.20	84.17	Chain-Chain
	0.40		
	0.50		
	0.61		
	0.80		
MN	0.20	84.25	Chain-Ring
	0.40		
	0.50		
	0.60		
	0.80		

cm³. The mixtures were prepared introducing first the less volatile component.

Equipment

The determination of the thermodiffusion coefficient in this work was done by the thermogravitational technique (Blanco et al. 2008). By this technique it is possible to determine the thermodiffusion coefficient from the variation of the concentration with the height of the column and some thermophysical properties of the analyzed mixture. The principle of the thermogravitational technique is as follows. Instead of trying to avoid convection in the elemental Soret cell, the thermogravitational technique adds it to the thermodiffusive separation. The liquid is inside a vertical gap with a high aspect ratio $L_z \gg L_x$ (where L_z is the height of the column and L_x is the width of the gap), and where the walls are at different and constant temperatures. This horizontal temperature gradient makes, generally, the denser component migrate to the cold wall due to thermodiffusion and at the same time, it is moved to the bottom of the column due to convection. Therefore, a vertical concentration gradient is established through the height of the gap. The theory of the thermogravitational column (Furry et al. 1939) allows

relating the stationary separation with the thermodiffusion coefficient, D_T , by the following expression:

$$\Delta c = -\frac{504L_z D_T \nu}{gL_x^4} c_0(1 - c_0) \tag{1}$$

where Δc is the difference of mass fraction between the top and the bottom of the gap, c_0 is the mass fraction of the initial mixture, $\alpha = -(1/\rho)(\partial\rho/\partial T)$ is the thermal expansion coefficient, ν is the kinematic viscosity and g is the gravity acceleration.

In this work we will take as reference component the denser component in each mixture. When $D_T > 0$ the denser component migrates to the cold wall and it is moved to the bottom of the column, increasing the concentration there, so that $\Delta c < 0$.

The thermogravitational column used in this work has parallelepiped configuration and it was developed and manufactured in Mondragon Goi Eskola Politeknikoa. It is shown in Fig. 1. The geometric parameters are: $L_z = 0.5 \pm 0.001$ m and $L_x = 1 \times 10^{-3} \pm 5 \times 10^{-6}$ m. The temperatures of the walls were $T_H = 301$ K and $T_C = 295$ K, which implies a mean temperature of 298 K. The column has four outlets of samples distributed evenly along the height of the column.

In order to determine the mass separation between the extremes of the column it was necessary to carry out a prior



Fig. 1 Thermogravitational column with parallelepiped configuration

calibration which relates the mass fraction with a physical property of the mixture, in our case, the density. An Anton Paar DMA 5000 vibrating quartz U-tube densimeter with accuracy of 5×10^{-6} g/cm³ was used to determine the density of the mixture at different concentrations. From these measurements the mass expansion coefficient may also be determined.

When stationary state is achieved, the vertical density gradient is determined $\partial\rho/\partial z$, by measuring the density of the samples extracted from the four outlets along the height of the column. In every case the variation of the density with the height of the column was linear. As example, Fig. 2 shows the variation of the density with the height for the system MN-nC16 with a 40 % of concentration of nC16.

The stationary separation between the extremes of the column is:

$$\Delta c = -\frac{L_z \partial\rho}{\beta \rho \partial z} \tag{2}$$

where β is the mass expansion coefficient defined by:

$$\beta = \frac{1 \partial\rho}{\rho \partial c} \tag{3}$$

Combining Eqs. 2 and 3 we can obtain:

$$D_T = -\frac{gL_x^4}{504} \cdot \frac{\alpha}{c_0(1 - c_0)\beta\mu} \cdot \frac{\partial\rho}{\partial z} \tag{4}$$

Where μ is the dynamic viscosity and $c_0(1 - c_0)$ is the product of the initial concentrations. This equation allows us determining D_T from the measurements of the density of the samples taken from different heights of the column. Considered the experimental errors committed in the measurements of the thermophysical properties, the estimated uncertainty in the determination of the thermodiffusion coefficient is of a 5 %.

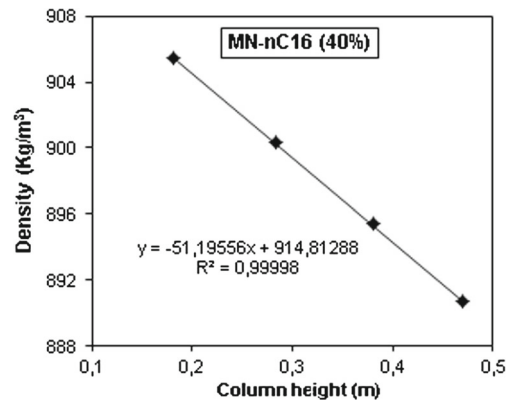


Fig. 2 Variation of the density of the mixture with the height of the column

The dynamic viscosity is determined by two viscometers: a HAAKE falling ball viscometer and a microviscometer Anton Paar AMVn.

Results and Discussion

In this section the results obtained in this work and the discussion generated around them are presented. The density, the mass expansion coefficient, the thermal expansion coefficient, the dynamic viscosity and the thermodiffusion coefficient have been determined for the 20 mixtures presented in the previous section. Table 2 shows the numerical values of these properties and of the mass separation for each mixture.

Figure 3 shows the variation of the density with the concentration of nC16 for the four studied systems. For the systems nC6-nC16 and nC10-nC16 this dependency is linear as it was expected, taking into account the quasi-ideality of the mixtures of n-alkanes. On the contrary, for the systems Tol-nC16 and MN-nC16 this dependency is quadratic, because they are non-ideal mixtures. It is interesting to point out that the four mixtures intersect in the point corresponding to the density of the nC16.

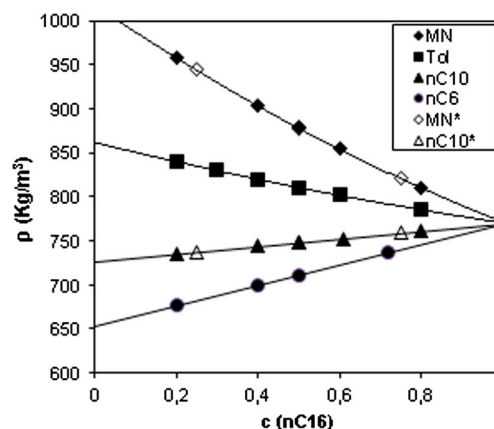


Fig. 3 Variation of the density with the concentration of nC16 for the four systems analyzed. Values with * correspond to the works (Leahy-Dios and Firoozabadi 2007; Leahy-Dios et al. 2008)

In Table 2, the values of β as obtained from the study of the density with the concentration of nC16 are also reported.

Figure 4 shows the results corresponding to the variation of the thermal expansion coefficient with the concentration. The variation of α with the concentration is quasi-linear in every case, and the four lines converge

Table 2 Results of the thermophysical properties, mass separation and thermodiffusion coefficient for the 20 mixtures analyzed in this work at 298 K where c is the mass fraction of nC16

System	c	ρ (kg/m ³)	$\alpha \times 10^{-3}$ (K ⁻¹)	β	μ (mPa·s)	$-\Delta c \times 10^{-2}$	$D_T \times 10^{-12}$ (m ² /sK)
Tol-nC16	0.80	785.15	0.929	0.101	1.77	—	—
Tol-nC16	0.60	801.76	0.962	0.108	1.20	—	—
Tol-nC16	0.50	810.65	0.980	0.112	1.02	1.14	1.30
Tol-nC16	0.40	820.00	0.998	0.116	0.87	1.26	1.86
Tol-nC16	0.30	829.78	1.017	0.120	0.76	1.35	2.63
Tol-nC16	0.20	840.00	1.039	0.125	0.68	1.12	3.54
MN-nC16	0.80	809.97	0.877	0.253	2.82	6.61	4.03
MN-nC16	0.60	854.11	0.846	0.273	2.69	9.61	4.19
MN-nC16	0.50	878.45	0.830	0.282	2.62	10.10	4.37
MN-nC16	0.40	903.28	0.810	0.288	2.65	9.88	4.42
MN-nC16	0.20	958.12	0.774	0.304	2.73	6.94	4.66
nC6-nC16	0.72	736.56*	1.009*	0.162*	1.26*	5.55*	6.38*
nC6-nC16	0.50	708.00	1.110	0.161	0.74**	5.08	8.37
nC6-nC16	0.40	699.21	1.140	0.161	0.56	4.16	9.60
nC6-nC16	0.20	676.84	1.233	0.159	0.40	2.18	11.10
nC10-nC16	0.80	761.07	0.917	0.058	2.13	2.39	1.90
nC10-nC16	0.61	752.85*	0.952*	0.059*	1.76*	3.33*	2.23*
nC10-nC16	0.50	747.77	0.966	0.059	1.52**	3.33	2.47
nC10-nC16	0.40	743.66	0.980	0.058	1.28	2.93	2.71
nC10-nC16	0.20	735.00	1.002	0.058	0.99	1.77	3.18

* Blanco et al. (2008)

** Alonso de Mezquia et al. (2012)

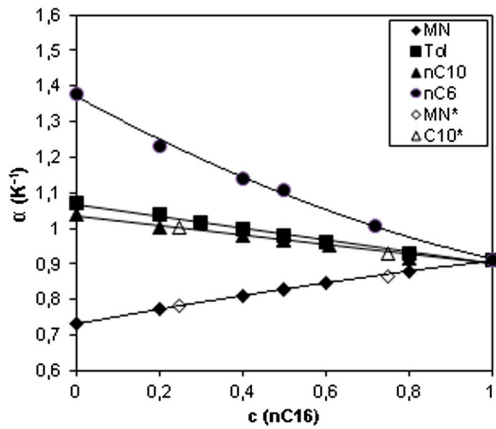


Fig. 4 Variation of the thermal expansion coefficient with the concentration of nC16 for the four mixtures analyzed. Values with * correspond to the works (Leahy-Dios and Firoozabadi 2007; Leahy-Dios et al. 2008)

to the value corresponding to a 100 % concentration of nC16.

Figure 5 shows the dependency of the dynamic viscosity with the composition of the mixture. In the same way as in the case of density and thermal expansion coefficient, the values of the viscosities in the four systems converge to the value of the viscosity of hexadecane when the compositions of the mixtures tend to 100 %.

In Figs. 3–5 are represented also the values obtained in Leahy-Dios and Firoozabadi (2007) and Leahy-Dios et al. (2008) for these thermophysical properties. The agreement between our values and the ones of those works can be considered excellent.

Moreover, Fig. 6 shows the experimental results obtained in our column for the separation, Δc , for the different mixtures of the four systems. These results are the mean value of at least three different measurements.

With the values of ρ , α , μ and Δc obtained in this work and the geometric constants of our thermogravitational

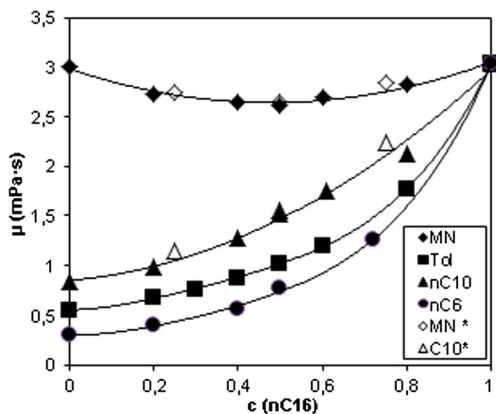


Fig. 5 Variation of the dynamic viscosity with the concentration of nC16 for the four systems analyzed. Values with * correspond to the works (Leahy-Dios and Firoozabadi 2007; Leahy-Dios et al. 2008)

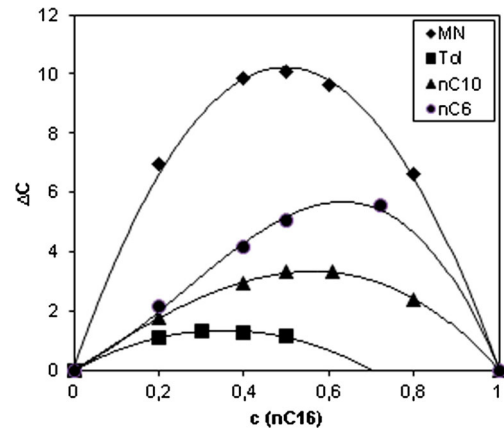


Fig. 6 Variation of the separation along the height of the column, with the concentration of nC16 in the four systems analyzed

column, the thermodiffusion coefficients can be calculated by Eq. 4. Figure 7 shows the obtained values of D_T . All the values of D_T are positive, which means that in every case the denser and the less dense components enrich the bottom and the top of the column respectively. It is important to point out that with our experimental equipment it has not been possible to measure thermodiffusion coefficients for the mixture Tol-nC16 with concentrations of nC16 higher than 50 %. In these mixtures the separation is very small and the experimental error masks the result. However, the behaviour of D_T in these systems indicates that for concentrations of nC16 higher than 65 % negative values of D_T can be achieved. In any case and for the four systems, the values of D_T decrease with the concentration of nC16, therefore, they decrease with the concentration of the heaviest component.

Considering Fig. 7 it can be observed that the dependency of D_T with the composition is linear for the four systems. Madariaga et al. (2010) showed that the behaviour of the thermodiffusion coefficient with the mass fraction is linear

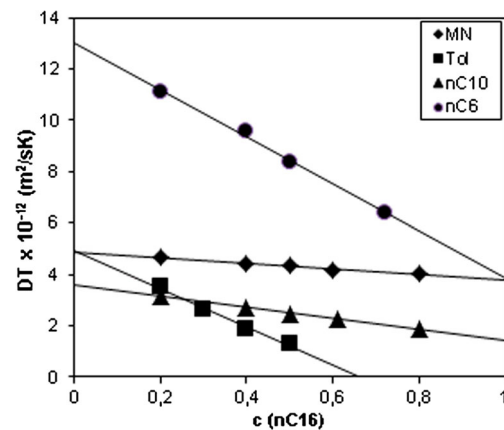


Fig. 7 Variation of the thermodiffusion coefficient with the concentration of nC16 for the four mixtures analyzed

in binary mixtures of n-alkanes. In this work we can confirm this behaviour for the systems nC6-nC16 and nC10-nC16. In this work also, we have shown that this linear behaviour appears also in non-ideal mixtures formed by linear chains and rings such as Tol-nC16 and MN-nC16.

It has to be pointed out that the coefficients D_T for the systems Tol-nC16 and MN-nC16 have the same value when the mass fraction of the hexadecane tends to zero, although their thermophysical properties and molecular weights are different. In other words, toluene and 1-methylnaphtalene have the same response to temperature gradient when hexadecane is present as traces, indicating that the mobility of these components is similar.

With regard to the slopes of the lines that represent the dependency of D_T with mass fraction, the four systems can be classified in two groups. One of the groups is formed by the systems nC6-nC16 and Tol-nC16 which have similar slopes, and the other group is formed by MN-nC16 and nC10-nC16 with similar slopes between them but different to the previous ones. Each group has in common that the difference of molecular weight between their components, ΔM , has similar values (see Table 1). In other words, the slopes of the lines D_T vs c are directly related to the values of ΔM .

The thermodiffusive behaviour of the systems formed by n-alkanes, nC6-nC16 and nC10-nC16, is perfectly described by the empirical relations obtained in Madariaga et al. (2010). In this work it is concluded that the values of the coefficient D_T are directly proportional to the difference of molecular weights and to the thermal expansion coefficient, and inversely proportional to the dynamic viscosity (Eq. (5) in Madariaga et al. (2010)). Therefore, for binary mixtures of n-alkanes it is satisfied that:

- When the difference between the molecular weights of the components increases, the mobility of the molecules increases, and therefore, the thermodiffusion coefficient increases too.
- When the dynamic viscosity increases the mobility of the molecules decreases, and the thermodiffusion coefficient decreases too.
- When the thermal expansion coefficient increases, the thermodiffusion coefficient increases too.
- The similarity between the molecules decreases the thermodiffusion coefficient. This is because molecules with the same shape respond in the same way to thermal effects, and therefore, the separation due to the thermodiffusive effect is smaller (Leahy-Dios and Firoozabadi 2007).

It can be observed how when comparing the systems nC6-nC16 and nC10-nC16, the previous considerations about ΔM , viscosity and thermal expansion coefficient encourage

the system nC6-nC16 and this is the one which has higher thermodiffusion coefficient (Fig. 6).

On the contrary, the behaviour of the thermodiffusion coefficient comparing systems with an aromatic ring is just the opposite. In these systems, as Fig. 7 shows, the thermodiffusion coefficient increases when viscosity increases and difference between molecular weights of the components and thermal expansion coefficient decrease. For example, the mixture MN-nC16 with a 50 % of mass fraction which has a viscosity of 2.63 mPa·s, has a D_T of 4.37 m²/sK; however, the system Tol-nC16 with a 50 % of mass fraction, which has a viscosity of 1.02 mPa·s, has a D_T of 1.30 m²/sK, which is a behaviour completely opposite to the one followed by systems of n-alkanes. A similar situation is found when the influence of α is studied. For the mixture Tol-nC16 with a 50 % of mass fraction α is 0.980 K⁻¹ and it has a D_T of 1.30 m²/sK; on the contrary, the mixture MN-nC16 with a 50 % of mass fraction has a α of 0.830 K⁻¹ and a D_T of 4.37 m²/sK.

Leahy-Dios and Firoozabadi (2007) have disclosed the influence of the molecular shape on the thermodiffusion coefficient. These authors, by experimental measurement of D_T in the series MN-nC_i and nC10-nC_i (i = 5-16) conclude that D_T values are higher in systems formed by MN and n-alkanes than in the ones formed by two n-alkanes. The normal alkanes have a similar configuration and therefore, they respond in the same way to the established thermal gradient, which would explain the low values of D_T in these systems compared to the ones obtained for MN-nC_i. Overall, these conclusions are not maintained for our results. The value of D_T for the system MN-nC16 is higher than the one obtained for the system nC10-nC16, the two systems with similar ΔM . However, for the systems Tol-nC16 and nC6-nC16 with similar values of ΔM the situation is reversed; the value of D_T for the system nC6-nC16 is much higher than the one obtained for the system Tol-nC16. In any case, more studies are needed to confirm the suggestion of Leahy-Dios and Firoozabadi (2007) about the importance of the molecular shape in the thermodiffusion coefficient.

On the other hand, these authors associate the mobility of each component to the Brownian movement of the molecules and it is, ultimately, a function of the viscosity. They also associate the similarity between the components of the mixture to the type of answer to a given force field. In our work, we have checked that in n-alkane mixtures it effectively happens that when molecular weight and viscosity decreases, D_T increases; but in the mixtures of Tol-nC16 and MN-nC16 the behaviour is just the opposite. This different behaviour can be caused by a strong influence of the similarity between the components which counteracts the previous effect, or by other properties that have not been studied in this work.

Conclusions

In this work the thermophysical properties (density, dynamic viscosity and mass and thermal expansion coefficients) and the thermodiffusion coefficient of 20 binary mixtures have been measured. All the mixtures have a common component (hexadecane) and the other component is a n-alkane (hexane or decane) or an aromatic ring (toluene or 1-methylnaphthalene).

For the four studied systems hexane-hexadecane, decane-hexadecane, toluene-hexadecane and 1-methylnaphthalene-hexadecane, it has been shown a linear dependency between the thermodiffusion coefficient and the mass fraction of the mixture. In addition, in the mixtures of hexane and decane, the behaviour is the one predicted by the empirical expressions of Madariaga et al. (2010) for n-alkane binary mixtures. In these mixtures the thermodiffusion coefficient is proportional to the difference of molecular weights of the components and to the quotient between the thermal expansion coefficient and the viscosity. On the contrary, for mixtures where one of the components has an aromatic ring, the behaviour is the opposite; it is proportional to the quotient between the viscosity and the thermal expansion coefficient.

More experiments in non-ideal binary mixtures are needed in order to clarify the influence of different parameters and to determine general trends in these binary mixtures. The knowledge about the behaviour of the thermodiffusion coefficient in binary mixtures will allow having a better basis for the study of ternary mixtures.

Acknowledgments The results presented in this work were obtained in the framework of the following projects: GOVSORET3 (PI2011-22), MICROSCALE (IE13-360), Research Groups (IT557-10), and Research Fellowship (BFI-2011-295) of Basque Government and DCMIX (DCMIX-NCR-00022-QS) from the European Space Agency.

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