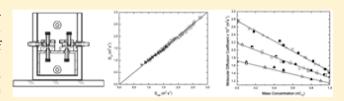
Determination of Molecular Diffusion Coefficient in n-Alkane Binary **Mixtures: Empirical Correlations**

D. Alonso De Mezquia, M. Mounir Bou-Ali, M. Larrañaga, J. A. Madariaga, and C. Santamaría

ABSTRACT: In this work we have measured the molecular diffusion coefficient of the *n*-alkane binary series nC_i-nC₆, nC_inC₁₀, and nC_i-nC₁₂ at 298 K and 1 atm and a mass fraction of 0.5 by using the so-called sliding symmetric tubes technique. The results show that the diffusion coefficient at this concentration is proportional to the inverse viscosity of the mixture. In addition, we have also measured the diffusion



coefficient of the systems nC_{12} - nC_{6} , nC_{12} - nC_{7} , and nC_{12} - nC_{8} as a function of concentration. From the data obtained, it is shown that the diffusion coefficient of the n-alkane binary mixtures at any concentration can be calculated from the molecular weight of the components and the dynamic viscosity of the corresponding mixture at 50% mass fraction.

I. INTRODUCTION

A concentration gradient within a mixture results in a mass transport of its components from the zones of a higher concentration to those of a lower concentration. This phenomenon, known as molecular diffusion, has generated great interest since its discovery in the nineteenth century when, first in gases and later on in liquids, it was studied with the aim of understanding the atom's behavior.

It was in 1855 when Fick established the first quantitative relation for the molecular diffusion phenomenon, known nowadays as Fick's law. Since then, interest in this transport property has increased, discovering a great number of fields in which the molecular diffusion coefficient has a huge influence, such as medicine or phisiology.²

As a consequence of this interest, there have appeared a great number of apparatus designed for the determination of the molecular diffusion coefficient. There are, for example, techniques that employ the principle of Taylor dispersion³ or other techniques such as open ended capillary (OEC)⁴ or thermal diffusion forced Rayleigh scattering (TDFRS)⁵ developed for the study of the molecular diffusion coefficient in liquid mixtures. The different techniques used for the determination of the molecular diffusion coefficient can be found elsewhere. 2,6-9

Some of these techniques, such as the OEC, have already been satisfactorily used in several studies earlier. 4,10 With the aim of correcting some of the limitations of this technique, the sliding symmetric tubes (SST) technique has been developed. This new technique takes out perturbations that appeared in the OEC technique, especially in aspects such as evaporations or manipulation of the apparatus, reducing, at the same time, the liquid quantity needed to carry out a measurement. In this work a complete description of the SST technique will be done, including the improvements obtained compared with the OEC

technique, and the validation process used to validate this new technique.

On the other hand, nowadays, there exist several models developed for the estimation of the molecular diffusion coefficient. 11,12 In order to check the validity of these models, it is necessary to compare the theoretical values obtained with the experimental ones. On some occasions, the procedure that checks these models requires a huge number of experimental values, which, at the same time, requires a great experimental effort. In this work, a new correlation, which is capable of predicting the molecular diffusion coefficient in normal alkane binary mixtures, is presented. The use of this correlation may help in the development of new theories or could be useful in verifying the existing ones.

This article is organized as follows: In section II, the SST technique is presented, and a complete description of the experimental installation and its analytical resolution is done. In this section, the process used to validate the technique is also included. In section III, the results obtained in the measurement of the molecular diffusion coefficient in several normal alkane mixtures are presented, to continue with the discussion and the development of new correlations for the prediction of the molecular diffusion coefficient (D_{12}) in normal alkane mixtures. In section IV, the conclusions obtained are based on the study carried out in this work.

II. SST TECHNIQUE

A. SST Technique Description. The SST technique consists of several sets of two identical vertical tubes. Each set has two positions: "faced tubes" (Figure 1a), in which the

Received: October 13, 2011 Revised: January 12, 2012 Published: January 20, 2012

[†]Mechanical and Manufacturing Department, Engineering Faculty, Mondragon Unibertsitatea, Loramendi 4 Apdo. 23, 20500 Mondragon, Spain

^{*}Department of Applied Physics II, University of Basque Country, Apdo. 644, 48080 Bilbao, Spain

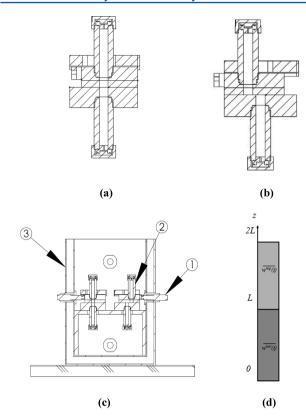


Figure 1. (a) Scheme of the sets of the SST technique on "faced tubes" configuration. (b) Scheme of the sets of the SST technique on "separated tubes" configuration. (c) Scheme of the SST sets (2) inside the water bath (3) in both their configurations: "faced tubes" position (left) and "separated tubes" position (right) and the screws (1) used to make the tubes slide. (d) Scheme of SST technique. $\overline{w^{\text{top}}}(t)$ and $\overline{w^{\text{bot}}}(t)$ are the mass fractions of component i in the top and bottom recipient, respectively.

mass transfer between both tubes is permitted, and "separated tubes" (Figure 1b). In this position, the content of the tubes is isolated. At the beginning of an experiment, the studied mixture with a slight difference of mass fraction of its components is introduced in each of the tubes. The mixture with a higher mass fraction of the denser component is introduced in the bottom tube, and the upper tube is filled with the mixture with a less concentration of the denser component so that the convective instability is avoided.

The SST technique introduces some improvements compared with the OEC technique on which it is based. For example, since the mixture is contained inside closed tubes, evaporations are eliminated, which gives the advantage of using this technique with volatile mixtures without reducing its efficiency. Another fact that has been improved has been the mixture quantity needed to carry out an essay. The OEC technique requires a huge quantity of mixture (400 cm³ approximately). This quantity is highly reduced in the SST technique, as it only needs around 100 cm³ of mixture in each experiment. One more improvement has been done related to the setup handling. In the OEC technique, whenever it is needed to analyze the experiment evolution, it is necessary to manipulate the setup. This fact affects the results of the experiment, which are highly influenced by the experimenter's expertise. However, in the SST technique, thanks to its design, all of these influences have been eliminated.

B. Analytical Solution. Assuming Fick's second law in one dimension (*z*: vertical direction), the following can be written:

$$\frac{\partial w}{\partial t} = D_{12} \frac{\partial^2 w}{\partial z^2} \tag{1}$$

where D_{12} is the molecular diffusion coefficient, which is considered constant for small mass fraction (w) changes. The formulation can be implemented as can be seen in Figure 1d considering the following boundary conditions:

$$w(z, 0) = \begin{cases} w^{\text{bot}} & 0 \le z < L \\ w^{\text{top}} & L < z \le 2L \end{cases}$$
 (2)

$$\frac{\partial w(z, t)}{\partial t} = 0 \text{ at } z = 0, \ 2L \text{ and } t > 0$$
(3)

where w^{bot} and w^{top} are the mass fractions of the bottom and upper tube, respectively, and L is the tube length. Solving eqs 1–3, we have

$$w(z,t) = \frac{w^{\text{top}} + w^{\text{bot}}}{2} - \frac{2(w^{\text{top}} - w^{\text{bot}})}{\pi}$$
$$\sum_{n=0}^{\infty} \frac{(-1)^n e^{\phi} \cos\left(\frac{2n+1}{2}\pi \frac{z}{L}\right)}{2n+1}$$
(4)

where $\varphi = -((2n + 1)/2)^2(\pi/L)^2D_{12}t)$. Therefore, at the middle point between both the tubes, we have

$$w(L, t) = \frac{w^{\text{top}} + w^{\text{bot}}}{2} \tag{5}$$

The change of mass fraction in each tube in function of time can be obtained using the following equations:

$$\overline{w^{\text{top}}(t)} = \frac{1}{L} \int_{L}^{2L} w(z, t) dz
= \frac{w^{\text{top}} + w^{\text{bot}}}{2} + \frac{4(w^{\text{top}} - w^{\text{bot}})}{\pi^{2}}
\sum_{n=0}^{\infty} \frac{e^{\Phi}}{(2n+1)^{2}}
\overline{w^{\text{bot}}(t)} = \frac{1}{L} \int_{0}^{L} w(z, t) dz
= \frac{w^{\text{top}} + w^{\text{bot}}}{2} - \frac{4(w^{\text{top}} - w^{\text{bot}})}{\pi^{2}}
\sum_{n=0}^{\infty} \frac{e^{\Phi}}{(2n+1)^{2}}$$
(6)

where $\overline{w^{\text{top}}}$ and $\overline{w^{\text{bot}}}$ are the mean mass fractions in the upper and bottom tube, respectively. The solution of the inverse problem posed by eqs 6 and 7 to obtain the molecular diffusion coefficient (D_{12}) is solved by the least-squares method using Matlab software. Solving expressions 6 and 7 when n=0, an initial guess of the molecular diffusion coefficient D_{12} is obtained. This initial value is then used for obtaining the D_{12} coefficient iterating the eqs 10 times and using terms on the summation up to n=1000. The more experimental points used, the more accurate the estimated value of the molecular

diffusion coefficient. Typically up to 11 points are used in each experiment. This tactic allows the use of experimental points from the very beginning of the experiment, which does not happen in other similar techniques that require some initial experimental time before beginning the analysis of experimental data.^{3,8}

C. Equipment and Procedure. We have used the SST equipment designed and constructed in Mondragon Goi Eskola Politeknikoa and described elsewhere. The two tubes with which each set of the device is provided have the same dimensions (length of 60 ± 0.01 mm and diameter of 9 ± 0.01 mm).

The experimental procedure starts with the preparation of two mixtures of 50 cm³ with a mass fraction difference (± 4 –6 wt %) so that the mean concentration of both mixtures corresponds with the one of the mixture to be studied. The tubes of several sets (typically 10) in their "separated tubes" position are filled with the mixtures, next introducing the sets in the water bath (Figure 1d) so that the mixture obtains the working temperature (T=298 K). The temperature of the water in this bath is controlled by a thermostatic bath, which has a temperature control of 0.1 K. In order to make sure that the studied mixture is in the working temperature, the sets in their "separated tubes" configuration are introduced in the bath no less than 48 h before the beginning of the experiment.

Once the mixture has reached the working temperature, all the sets in the bath are changed to their "faced tubes" configuration, starting the diffusion process in this manner. With the aim of making this transition, the bath has some external screws (Figure 1c) that make sliding of the tubes possible in a controlled way.

In determined time intervals, the position of the sets is changed one by one using the screws back to the "separated tubes position." The time intervals in which the set's positions are changed are between 6, 12, or 24 h, depending on the speed at which sensitive concentration changes occur in the tubes. After stopping the mass transfer process between the tubes, the sets are extracted from the bath, and the concentration in each of them is analyzed. This allows obtaining the concentration change in the tubes in function of time.

The SST technique was validated ^{13,14} using five binary mixtures: the three binary mixtures composed of pairwise combinations of tetrahydronaphtalene, normal dodecane, and isobutylbenzene at 298 K and 50 wt % of each component used in a benchmark test ¹⁶ to compare various experimental techniques and the binary mixtures: water—ethanol at 298 K and 60.88 wt % water and toluene-n hexane at 298 K and 51.67 wt % toluene, which have been widely studied in the literature. ^{17–20} The experimental test was repeated at least twice. The reproducibility of the experimental results of the SST technique is within 3% of deviation. In general, the differences with published data are, on an average, below 3%.

The determination of the mass fraction of a mixture is essential to determine molecular diffusion coefficients. In binary mixtures, the mass fraction is related to one physical property of the mixture (the density in this experimental study). An Anton Paar DMA 5000 vibrating quartz U-tube densimeter having an accuracy of $\pm 5\times 10^{-6}~\text{g/cm}^3$ is used to determine the density of the mixture. Since we are working with small mass fraction differences, it can be supposed that

$$\rho = aw_i \tag{8}$$

where ρ is the density of the mixture, w_i is the mass fraction of component i in the binary mixture, and a is a constant

Table 1. Values of Viscosity (μ) and Molecular Diffusion Coefficient (D_{12}) for the nC₆-nC_i, nC₁₀-nC_i, and nC₁₂-nC_i Series at 50 wt % and 298 K^a

system	μ mPa s	$D_{12} 10^{-9} \text{ m}^2 \text{ s}^{-1}$	$(\overline{D_{12}})_{cc}10^{-9} \text{ m}^2 \text{ s}^{-1}$	δ (%)					
set 1: nC _i -nC ₆									
nC_{10} - nC_{6}	0.4704	2.53	2.51	0.8					
nC_{11} - nC_{6}	0.5312	2.27	2.22	2.1					
nC_{12} - nC_6	0.5581	2.09	2.11	-1.2					
nC_{13} - nC_{6}	0.6103	1.96	1.93	1.4					
nC_{14} - nC_{6}	0.6556	1.76	1.80	-2.3					
nC_{15} - nC_{6}	0.6938	1.62	1.68	-3.7					
nC_{16} - nC_{6}	0.7446	1.54	1.58	-2.6					
nC_{17} - nC_6	0.7737	1.46	1.52	-4.1					
nC_{18} - nC_{6}	0.8265	1.39	1.43	-2.9					
set 1: nC_i - nC_{10}									
nC_5 - nC_{10}	0.3993	3.03	2.96	2.3					
nC_6 - nC_{10}	0.4704	2.54	2.51	1.2					
nC_7 - nC_{10}	0.5633	2.09	2.09	-0.0					
$nC_{14}-nC_{10}$	1.2929	0.913	0.915	-0.2					
$nC1_5$ - nC_{10}	1.3972	0.819	0.845	-3.1					
nC_{16} - nC_{10}	1.5150	0.756	0.779	-3.0					
$nC_{17}-nC_{10}$	1.6337	0.702	0.722	-2.8					
nC_{18} - nC_{10}	1.7780	0.656	0.664	-1.2					
set 1: nC_{i} - nC_{12}									
nC_5 - nC_{12}	0.4551	2.57	2.59	-0.9					
nC_6 - nC_{12}	0.5581	2.09	2.11	-1.2					
nC_7 - nC_{12}	0.6823	1.70	1.73	-1.7					
nC_8 - nC_{12}	0.8071	1.49	1.46	1.9					
nC_9 - nC_{12}	0.9452	1.24	1.25	-0.8					
nC_{16} - nC_{12}	2.0335	0.589	0.580	1.5					
nC_{17} - nC_{12}	2.2323	0.547	0.529	3.2					
$nC_{18}\text{-}nC_{12}$	2.3945	0.503	0.493	2.0					

 $^a(\overline{D_{12}})_{cc}$ is the diffusion coefficient calculated from eq 9, and δ is the relative deviation between D_{12} and D_{cc} .

Table 2. Diffusion Coefficient (D_{12}) in the nC_{12} - nC_6 , nC_{12} - nC_7 , and nC_{12} - nC_8 Systems for Different Mass Fractions of nC_{12} (w_1) at 298 K^a

mixture	w_1	$D_{12} \ 10^{-9} \ \mathrm{m^2 \ s^{-1}}$	$(D_{12})_{\rm cc}~10^{-9}~{\rm m}^2~{\rm s}^{-1}$	δ (%)
nC_{12} - nC_{6}	0.18	2.48	2.55	-3.0
	0.46	2.24	2.17	3.1
	0.50	2.13	2.11	0.7
	0.66	1.93	1.88	2.2
	0.82	1.72	1.67	2.9
	0.95	1.51	1.49	1.0
nC_{12} - nC_{7}	0.16	2.09	2.04	2.6
	0.42	1.84	1.80	2.2
	0.50	1.70	1.73	-1.7
	0.63	1.64	1.61	1.6
	0.80	1.48	1.46	1.2
	0.94	1.37	1.33	2.9
nC_{12} - nC_{8}	0.10	1.70	1.69	0.6
	0.40	1.56	1.52	2.5
	0.50	1.49	1.46	2.0
	0.80	1.25	1.28	-2.4

 $^a(D_{12})_{\rm cc}$ is the diffusion coefficient calculated from eq 19, and δ is the relative deviation between calculated and measured diffusion coefficient.

parameter. To determine the constant parameter, *a*, five mixtures of known concentration are prepared by weighing and then, the density of each mixture is measured. Thus, we

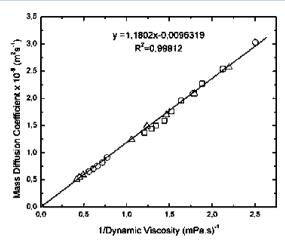


Figure 2. Diffusion coefficient as a function of the inverse of the dynamic viscosity for the systems nC_6 - nC_i (\square), nC_{10} - nC_i (\bigcirc), and nC_{12} - nC_i (\triangle), at 50 wt % and 298 K.

determined the mass fraction with a deviation around wt % \pm 0.01.

The dynamic viscosity (μ) of the studied mixtures has been measured with an Anton Paar AMVn falling ball Microviscosimeter with reproducibility better than 0.1%. All the measurements were done at 298 K.

III. RESULTS AND DISCUSSION

A. Studied Mixtures. All of the products used in this study were purchased from Merck with a purity better than 99%. The studied mixtures have been prepared by weight using a Gram VXI-310 digital scale with a precision of 0.001 g. and introducing first the less volatile component and then the corresponding amount of the second component; the mixture is finally shaken to ensure homogeneity. After the preparation, the density of the mixture was measured to confirm that the composition deduced from the calibration curve coincides with the prepared one. This was always the case.

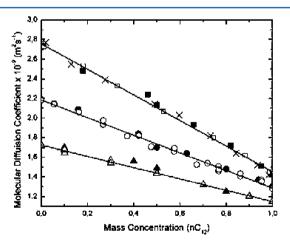


Figure 3. Diffusion coefficient as a function of the mass fraction of the denser component (nC_{12}) for the mixtures, nC_{12} - nC_6 , nC_{12} - nC_7 , and nC_{12} - nC_8 at 298 K. ■, nC_{12} - nC_6 this work; □, nC_{12} - nC_6 ref 22; ×, nC_{12} - nC_6 ref 24; ●, nC_{12} - nC_7 this work; ○, nC_{12} - nC_7 ref 23; ♠, nC_{12} - nC_8 this work; △, nC_{12} - nC_8 ref 21. The continuous lines represent a linear fit to the data.

In this study, 25 mixtures at 50 wt % have been prepared corresponding to the *n*-alkane series

$$nC_6 - nC_1 i = 10, 11, 12, 13, 14, 15, 16, 17, 18$$

$$nC_{10} - nC_{i}i = 5, 6, 7, 14, 15, 16, 17, 18$$

Table 3. Values of the Limiting Diffusion Coefficient (D_1^0 and D_2^0) Taken from Refs 3 and 23^a

system	D_1^0	D_2^0	D_1^0/D_2^0	M_1/M_2	$\delta(\%)$
nC_{16} - nC_{6}	2.21	0.85	2.60	2.62	0.7
nC_{12} - nC_{6}	2.74	1.42	1.93	1.97	2.5
nC_{12} - nC_{7}	2.19	1.30	1.69	1.70	0.6
nC_{12} - nC_{8}	1.71	1.15	1.49	1.49	0.0
$nC_{16}-nC_{12}$	0.67	0.49	1.36	1.33	-2.2
$nC_{7-}nC_{8}$	2.80^{b}	2.43 ^b	1.15	1.14	-1.7

 aM_1 and M_2 are the molecular masses of the components, and δ is the relative deviation between the ratios D_1^0/D_2^0 and M_1/M_2 . ^bInterpolated from ref 3.

$$nC_{12} - nC_i i = 5, 6, 7, 8, 9, 16, 17, 18$$

The measured values at 298 K for dynamic viscosity (μ) and mass diffusion coefficients for these mixtures are summarized in Table 1.

In addition, this study has been completed measuring the molecular diffusion coefficient of the systems nC_{12} - nC_6 , nC_{12} - nC_7 , and nC_{12} - nC_8 at 298 K at different concentrations. For this end, the following mixtures were prepared:

$$nC_{12} - nC_6 w_1 = 0.18, 0.46, 0.50, 0.66, 0.82, 0.95.$$

$$nC_{12} - nC_7 w_1 = 0.16, 0.42, 0.50, 0.63, 0.80, 0.94.$$

$$nC_{12} - nC_8 w_1 = 0.10, 0.40, 0.50, 0.80.$$

where w_1 is the mass fraction of dodecane.

In Table 2, the measured values of the diffusion coefficients of these systems at the concentrations considered are shown.

These systems were chosen, as hydrocarbon mixtures are quite regular in their properties. It was hoped that at least some general and useful empirical relationship would be found.

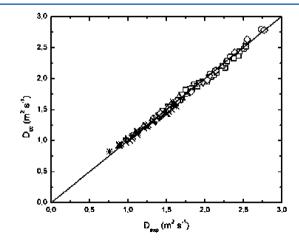


Figure 4. Experimental diffusion coefficient versus calculated from eq 19 diffusion coefficient. ×, nC_{10} - nC_7 ref 23; ○, nC_{12} - nC_6 ref 24; ○, nC_{12} - nC_6 ref 24; ○, nC_{12} - nC_7 ref 23; △, nC_{12} - nC_8 ref 21; +, nC_{14} - nC_7 ref 23; ×, nC_{14} - nC_8 ref 23; *, nC_{16} - nC_7 ref 22.

B. Correlation between Diffusion and Viscosity. In what follows, we shall analyze the relation between the diffusion coefficient and the dynamic viscosity for mixtures at a mass fraction equal to 50 wt % (equimass mixtures).

In Figure 2, the values of D_{12} as a function of the inverse of dynamic viscosity for the equimass mixtures in Table 1 are represented. As can be seen, the data fit well to a straight line, which passes through the origin and, thus, the following linear relation can be written:

$$\overline{D_{12}} = k \frac{1}{\overline{\mu}} \tag{9}$$

where $\overline{D_{12}}$ and $\overline{\mu}$ are the diffusion coefficient and the dynamic viscosity at $w_1 = 0.5$, respectively. The proportionality constant k is $k = 1.18 \times 10^{-12}$ kg m/s².

In Table 1, the values of $\overline{D_{12}}$ calculated using eq 9 for each mixture are also shown and in the last column, the relative deviations with the measured values are given. As can be seen, these deviations are smaller than 3% in all the cases. Thus, we can conclude that the product $\overline{D_{12}}\mu$ for equimass mixtures is a universal constant for n-alkane mixtures.

D. Dependence of the Diffusion Coefficient with Composition. According to Darken equation for thermodynamic ideal mixtures, the diffusion coefficient should be a linear function of the molar fraction. However, as was suggested in an earlier work by van Geet and Adamson,²¹ the diffusion coefficient in n-alkane mixtures is linear with the mass fraction but not with the mole fraction. To closely analyze the diffusion composition dependence we have measured the diffusion coefficient as a function of composition for the three systems: nC_{12} - nC_{6} , nC_{12} nC₇, and nC₁₂-nC₈. The results obtained appear in Table 2. In Figure 3, these data as a function of the mass fraction of the heavier component, w₁, are plotted. As can be seen, the ordinary diffusion coefficient is for each system a linear function of the mass fraction and decreases with the concentration of the heavier component. In this figure, the diffusion data in the literature for the systems considered $^{21-24}$ are also shown. These data coincide with our measurements within the experimental error.

Thus, the dependence of D_{12} with composition can be written as follows:

$$D_{12} = w_2 D_1^0 + w_1 D_2^0 (10)$$

where w_1 and w_2 are the mass fraction of heavier and lighter components, respectively, and D_1^0 and D_2^0 are the limiting diffusion coefficients defined by

$$D_1^0 = \lim_{w_1 \to 0} D_{12} \tag{11}$$

$$D_2^0 = \lim_{w_2 \to 0} D_{12} \tag{12}$$

In Table 3, the values of D_1^0 and D_2^0 for some selected mixtures reported in the literature^{3,23} covering a large range of mass ratio for liquid n-alkanes are given. As can be seen in this table, the ratio D_1^0/D_2^0 is always close to the ratio of the molecular masses M_1/M_2 of the two alkanes involved. Thus, we have

$$\frac{D_1^0}{D_2^0} = \frac{M_1}{M_2} \tag{13}$$

Introducing eq 13 in eq 10, one obtains

$$D_{12} = D_1^0 \left(w_2 + \frac{M_2}{M_1} w_1 \right) \tag{14}$$

For $w_1 = w_2 = 0.5$, the corresponding diffusion coefficient \overline{D}_{12} will be

$$\overline{D_{12}} = \frac{D_1^0}{2} \left(\frac{M_1 + M_2}{M_1} \right) \tag{15}$$

and then, eq 14 in terms of $\overline{D_{12}}$ can be written as

$$D_{12} = 2 \overline{D_{12}} \left(\frac{M_1 w_2 + M_2 w_1}{M_1 + M_2} \right) \tag{16}$$

In particular, the infinite dilution values $D_1^{\ 0}$ and $D_2^{\ 0}$ will be

$$D_1^0 = 2 \overline{D_{12}} \frac{M_1}{M_1 + M_2} \tag{17}$$

$$D_2^0 = 2 \overline{D_{12}} \frac{M_2}{M_1 + M_2} \tag{18}$$

Finally, using eq 9, the diffusion coefficient can be written as

$$D_{12} = \frac{2k}{\overline{\mu}} \left(\frac{M_1 w_2 + M_2 w_1}{M_1 + M_2} \right) \tag{19}$$

where $k = 1.18 \times 10^{-12} \text{ kg m/s}^2$.

Equation 16 allows determining D_{12} from $\overline{D_{12}}$ measurements, which, in turn, can be calculated using eq 9 from viscosity measurements.

To confirm the validity of this correlation, we have determined the values of D_{12} from eq 19 for the mixtures given in Table 2. As shown in the last column of this table, the relative deviations between these values and the experimental ones are inferior to the experimental error.

As an additional test of eq 19, we have taken the experimental D_{12} values as a function of concentration reported in the literature $^{21-24}$ for different n-alkane systems. In Figure 4, the values of D_{12} calculated using eq 19 are plotted against the corresponding experimental ones. As seen here, all data lie on a straight line through the origin with slope unity.

From what has been just mentioned, we can conclude that the diffusion coefficient of *n*-alkane mixtures at any concentration can be calculated from the data of dynamic viscosity for equimass mixtures.

IV. CONCLUSIONS

In this work, the diffusion coefficient of several *n*-alkane mixtures at 50 wt % has been measured by using the SST technique. From the results obtained, it is shown that the product of the diffusion coefficient and the dynamic viscosity for these mixtures is a universal constant independent of the alkane involved.

In addition, the diffusion coefficient of the systems nC_{12} - nC_{6} , nC_{12} - nC_{7} , and nC_{12} - nC_{8} in the whole concentration range has also been measured. We have obtained for each system a linear correlation between the diffusion coefficient and the mass fraction. This correlation allows determining the diffusion coefficient at any concentration from the molecular weight of the components of a mixture and the dynamic viscosity of the mixture at 50 wt %.

AUTHOR INFORMATION

Corresponding Author

*E-mail: mbouali@mondragon.edu.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This article presents results that were partly obtained in the framework of the following projects: GOVSORET3 (PI2011-22), MIBIO, and Research Groups (IT557-10) of Basque Government and DCMIX from the European Space Agency.

REFERENCES

- (1) Fick, A. Poggendorff's Ann. Phys. 1855, 94, 59.
- (2) Cussler, E. Diffusion Mass Transfer in Fluid Systems; Cambridge Univesity Press: New York, 1997.
- (3) Alizadeh, A. A.; Wakeham, W. A. Int. J. Themophys. 1982, 3 (4), 307.
- (4) Dutrieux, J.; Platten, J. K.; Chavepeyer, G.; Bou-Ali, M. M. J. Phys. Chem. B 2002, 106, 6104.
- (5) Wiegand, S.; Köhler, W. Thermal Nonequilib. Phenom. Fluid Mixt.; Springer: Berlin, 2002; pp 36-43.
- (6) Taylor, R.; Krishna, R. Multicomponent mass transfer; John Wiley & Sons Inc.: New York, 1993.
- (7) Cussler, E. Multicomponent diffusion; Elsevier Scientific Publishing Company: Amsterdam, 1976.
- (8) Rutten, P. W. M. Diffusion in liquids; Delft University Press: Delft, 1992.
- (9) Tyrrell, J. V. Harris, K. R. Diffusion in liquids; Butterworth & Co., Ltd.: London, 1984.
- (10) Leahy-Dios, A.; Bou-Ali, M. M.; Platten, J. K.; Firoozabadi, A. J. Chem. Phys. **2005**, 122, 234502.
- (11) Ghorayeb, K.; Firoozabadi, A. AIChE J. 2000, 46, 883-891.
- (12) Polyakov, P.; Müller-Plathe, F.; Wiegand, S. J. Phys. Chem. B 2008, 112, 14999.
- (13) Blanco P.; Bou-Ali M. M.; Urteaga P.; Alonso de Mezquia D.; Platten J. K. Sliding Symmetric Tubes: new technique for the molecular diffusion coefficients determination of liquid mixtures. 18th European Conference on Thermophysical Properties, Pau, France, 2008.
- (14) Alonso de Mezquia D.; Blanco, P.; Bou-Ali M. M.; Zebib A. New Technique for Measuring the Molecular Diffusion Coefficients of Binary Liquid Mixtures. *European Thermodynamics Seminar EURO-THERM* 84, Namur, Belgium, **2009**.
- (15) Blanco P., Bou-Ali M. M., Urteaga P.; Tubos Simétricos Deslizantes, Patente P200700132/2, 2008.
- (16) Platten, J. K.; Bou-Ali, M. M.; Costesèque, P.; Dutrieux, J.; Köhler, W.; Leppla, C.; Wiegand, S.; Wittko, G. *Philos. Mag.* **2003**, 83, 1965.
- (17) Leahy-Dios, A.; Firoozabadi, A. J. Phys. Chem. B 2007, 111, 191.
- (18) Zhang, K. J.; Briggs, M. E.; Gammon, R. W.; Sengers, J. V. J. Chem. Phys. **1996**, 104, 6881.
- (19) Kolodner, P.; Williams, H.; Moe, C. J. Chem. Phys. 1988, 88, 6512.
- (20) Köhler, W.; Müller, B. J. Chem. Phys. 1995, 103, 4367.
- (21) Van Geet, L. A.; Adamson, W. A. J. Phys. Chem. 1964, 68 (2), 238.
- (22) Bidlack, D. L.; Anderson, D. K. I. Phys. Chem. 1964, 68, 3790.
- (23) Lo, H. Y. Chem. Eng. Data Ser. 1974, 19 (3), 236-241.
- (24) Shieh, J. J. C.; Lyons, P. A. J. Phys. Chem. 1969, 73, 3258.