

Optimization of Taylor Dispersion Technique for Measurement of Mutual Diffusion in Benchmark Mixtures

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Abstract The Taylor dispersion technique has been used for measuring binary mutual diffusion coefficients for mixtures of 1,2,3,4-tetrahydronaphthalene (THN), isobutylbenzene (IBB) and dodecane ($C_{12}H_{26}$) at 0.5:0.5 mass fraction symmetric points, and for 0.9:0.1 mass fraction in IBB- $C_{12}H_{26}$. From the Stokes–Einstein equation and our experimental results, the limiting diffusion coefficients, D^0 , and the equivalent solvated radii, R_s , have been estimated at infinitesimal concentration of these species (TNH, IBB and $C_{12}H_{26}$). The measured diffusion coefficients are used to estimate activity coefficients of the components in the mixture, contributing to a better understanding of the structure of such systems and of their thermodynamic behaviour at different concentrations. We have also investigated the diffusion properties for a ternary system containing equal mass fractions of all the components (0.33THN: 0.33IBB: 0.33 $C_{12}H_{26}$) and at 298.15 K.

Keywords Diffusion · 1,2,3,4-tetrahydronaphthalene (THN) · Isobutylbenzene (IBB) · Dodecane ($C_{12}H_{26}$) · Taylor technique

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Introduction

The Taylor dispersion technique is a fast and reliable method for the measurement of mutual diffusion coefficients (D_{ik}) for binary and multicomponent solutions. It has been extensively tested for electrolytes and for dilute solutions e.g. Santos et al. (2015) and Ribeiro et al. (2011). Recently it has been extended to other systems of interest, such as the Fontainebleau benchmark mixture, which is a symmetrical mixture of 1,2,3,4-tetrahydronaphthalene (THN), isobutylbenzene (IBB) and dodecane ($C_{12}H_{26}$) at equal mass fraction (Gebhardt et al. 2013). This mixture is particularly important as it is widely used by the oil industry as a model (Shapiro et al. 2004) for understanding the thermodynamics inside hydrocarbon reservoirs during the exploration stage (Shapiro et al. 2004) This study is also motivated by the DCMIX program (Diffusion and Thermodiffusion Coefficients in Mixtures) of the European Space Agency (ESA), for which it was the first mixture investigated. Within the framework of this program (DCMIX1), experiments conducted on-board the International Space Station aim to measure the thermodiffusion effects in binary and ternary mixtures to validate results obtained on the ground, as reported in 2015 by Shevtsova et al. (2015), Mialdun et al. (2015), Galand and Van Vaerenbergh (2015), Ahadi and Saghir (2015) and Khlybov et al. (2015). Several international teams and laboratories have dedicated their work to the difficult task of establishing the isothermal diffusion coefficients for binary and ternary mixtures of these components. There is fair agreement between all the results reported in the literature, which have been obtained through various experimental methods, with deviations between them of the order of 8.5 % (Platten et al. 2003).

Table 1 Binary diffusion coefficients, D^a , for mixtures of tetralin (THN), isobutylbenzene (IBB) and dodecane ($C_{12}H_{26}$) at 298.15 K

Mass fraction w	$C_{12}H_{26}$ - IBB	$C_{12}H_{26}$ - THN	IBB - THN
0.5:0.5	0.898 ± 0.005	0.592 ± 0.010	0.786 ± 0.005
0.9:0.1	0.980 ± 0.050		

$$^a D_{ij} \pm S_{D_{ij}} / (10^{-9} \text{ m}^2 \text{ s}^{-1})$$

The main goal of this work is to adapt and optimize our Taylor dispersion equipment and protocol to the measurement of diffusion coefficients in the Fontainebleau benchmark mixture, which will provide additional trustworthy information on this benchmark mixture to the scientific community. This technique has established itself as a reliable experimental method for the determination of mutual diffusion coefficients in binary, ternary and even quaternary liquid systems (e.g. Santos et al. 2012, 2013, 2015; Ribeiro et al. 2006, 2010, 2011). For binary solutions, the usual procedure consists of the injection of a small volume of solution containing the solute at concentration $c \pm \Delta c$ into a laminar carrier stream of composition c . As the injected sample flows through a long capillary tube, radial diffusion and convection along the tube axis shape the initial δ concentration pulse into a Gaussian profile. Binary mutual diffusion coefficients (D) are calculated from the broadened distribution of the dispersed sample measured at the tube outlet. This technique has been extended to measure ternary mutual diffusion coefficients (D_{ik}) for multicomponent solutions. The results include diffusion cross-coefficients D_{ik} ($i \neq k$) which describe the coupled fluxes of solutes driven by concentration gradients in the other. Studies for binary mixtures involving 1,2,3,4-tetrahydronaphthalene,

isobutylbenzene and dodecane were made for 0.5–0.5 mass fraction symmetric points, and with IBB- $C_{12}H_{26}$ for 0.9:0.1 mass fraction. In case of ternary mixtures measures were made for liquids with equal mass fractions of all the components (0.33:0.33:0.33). The results obtained for all these systems at 298.15 K are presented in Tables 1 and 2.

Experimental

Materials

1,2,3,4-tetrahydronaphthalene (mass fraction purity 0.98, density 0.973 g cm^{-3}) and isobutylbenzene (mass fraction purity 0.99, density 0.850 g cm^{-3}) were supplied from Acros Organics. Dodecane (mass fraction purity 0.99, density 0.750 g cm^{-3}), was supplied by Panreac Synthesis. All liquids were used as received, with no further purification. Mixtures with various compositions were prepared by weighing each component using a Mettler H80 analytical balance with resolution 0.1 mg/160 g. Each mixture was freshly prepared and de-aerated for about 30 min before each set of runs.

Diffusion Measurements

The Taylor dispersion technique is well described in the literature as an experimental method for the measurement of diffusion coefficients, as in Tyrrel (1964), Barthel et al. (1996), Loh (1997), Alizadeh et al. (1980) and Callendar and Leait (2006). Only a brief description of the equipment and procedure used in the present study is presented here (see Santos et al. 2012, 2013, 2015; Ribeiro et al. 2006, 2010, 2011).

Table 2 Ternary diffusion coefficients, D_{ij}^a , for the mixture of THN (1), IBB (2) and $C_{12}H_{26}$ (0) at same mass fraction (i.e., 0.33:0.33:0.33) at 298.15 K

Mass fraction of each component in mixture w	D_{12} $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$	D_{12} $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$	D_{21} $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$	D_{22} $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$	\widehat{D}_1 $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$	\widehat{D}_2 $/(10^{-9} \text{ m}^2 \text{ s}^{-1})$
THN-IBB- $C_{12}H_{26}$						
(0.33):(0.33):(0.33)	1.004 ± 0.089 (−2.6 %)b (+4.0 %)c (−1.0 %)d	0.078 ± 0.046 (−76.3 %)b (−68.0 %)c (45.0 %)d	-0.448 ± 0.168 (2.6 %)b (+54.0 %)c (+12.3 %)d	0.560 ± 0.060 (−13.1 %)b (−8.5 %)c (−16.3 %)d	0.903 (−8.8 %)b	0.658 (−3.5 %)b

$$^a D_{ij} \pm S_{D_{ij}} / (10^{-9} \text{ m}^2 \text{ s}^{-1})$$

$$^b (D_{ij} - D_{ijLit}) / D_{ij} \%, \text{ where } D_{ijLit} \text{ represents the values obtained by Mialdun et al. (2013)}$$

$$^c (D_{ij} - D_{ijLit}) / D_{ij} \% \text{ where } D_{ijLit} \text{ are the average values calculated from data obtained by Mialdun et al. (2013) and Sechenyh et al. (2013)}$$

$$^d (D_{ij} - D_{ijLit}) / D_{ij} \% \text{ where } D_{ijLit} \text{ are the average values calculated from results published by Mialdun et al. (2013), Sechenyh et al. (2013) and Larrañaga et al. (2013)}$$

The Taylor method is based on the dispersion of small amounts of solution injected into laminar carrier streams of solution of different composition flowing through a long capillary tube. At the start of each run, a 6-port Teflon injection valve (Rheodyne, model 5020) was used to introduce 63 mm^3 of solution into a laminar carrier stream of slightly different composition. A flow rate of $0.20 \text{ cm}^3 \text{ min}^{-1}$ was maintained by a metering pump (Gilson model Minipuls 3) to give retention times of about $1.1 \times 10^4 \text{ s}$. The dispersion tube (length $32.799 (\pm 0.001) \text{ m}$) and the injection valve were kept at 298.15 K in an air thermostat. Dispersion of the injected samples was monitored using a differential refractometer (Waters model 2410) at the outlet of the dispersion tube. Detector voltages, $V(t)$, were measured at 5 s intervals with a digital voltmeter (Agilent 34401 A).

Binary diffusion coefficients were evaluated by fitting the detector voltages to the dispersion equation

$$V(t) = V_0 + V_1 t + V_{\max} (t_R/t)^{1/2} \exp[-12D(t - t_R)^2/r^2 t] \quad (1)$$

where r is the internal radius of the dispersion tube. The additional fitting parameters were the mean sample retention time t_R , peak height V_{\max} , baseline voltage V_0 , and baseline slope V_1 .

Mutual diffusion in ternary solutions is described by the equations

$$J_1 = -D_{11} \nabla c_1 - D_{12} \nabla c_2 \quad (2)$$

$$J_2 = -D_{22} \nabla c_1 - D_{21} \nabla c_2 \quad (3)$$

where J_1 , J_2 , ∇c_1 , and ∇c_2 are the molar fluxes and the gradients in the concentrations of solute 1 and 2, respectively. The main diffusion coefficients D_{11} and D_{22} give the flux of each solute produced by its own concentration gradient. Information about coupled diffusion is provided by cross-diffusion coefficients D_{12} and D_{21} . A positive D_{ik} cross-coefficient ($i \neq k$) indicates co-current coupled transport of solute i from regions of higher to lower concentrations of solute k . A negative D_{ik} coefficient indicates counter-current coupled transport of solute i from regions of lower to higher concentration of solute k .

The D_{ik} coefficients, defined by Eqs. 2 and 3 were evaluated by fitting the ternary dispersion Eq. 4

$$V(t) = V_0 + V_1 t + V_{\max} (t_R/t)^{1/2} \left[W_1 \exp\left(-\frac{12D_1(t - t_R)^2}{r^2 t}\right) + (1 - W_1) \exp\left(-\frac{12D_2(t - t_R)^2}{r^2 t}\right) \right] \quad (4)$$

The two pairs of refractive index profiles, D_1 and D_2 , are the eigenvalues of the matrix of the ternary D_{ik} coefficients. W_1 and $1 - W_1$ are the normalized pre-exponential factors.

In these experiments, small volumes, ΔV , of the solution of composition $c_1 \pm \Delta c_1$, $c_2 \pm \Delta c_2$, are injected into carrier solutions of composition, c_1 and c_2 at time $t = 0$. The concentrations of the injected solutions ($c \pm \Delta c$) and the carrier solutions (c), for both binary and ternary systems, differed by 10 % or less in mass. The mixtures in our experiments were prepared using mass fractions and later converted to molar concentrations (moles of solute per liter of solution) by means of the relation $w_i = c_i(M_i/\rho)$ where w_i stands for the concentration in mass fraction, c_i is molar concentration, M_i is the molar mass of the constituent i and ρ the density of the mixture.

Details have previously been reported of the method of calculation of the D_{ik} coefficients from the fitted values of D_1 , D_2 and W_1 by Deng and Leaist (1991). Following Legros et al. (2015), we have tested for the ternary mixture the smart strategy of tuning the injections so as to have the amplitude of one of the signals negative and obtain 'double dip' signals. The presence of these dissimilar shaped peaks demonstrates the existence of two modes, corresponding to different eigenvalues of the system, allowing to distinguish between them and avoiding the selection of inaccurate solutions.

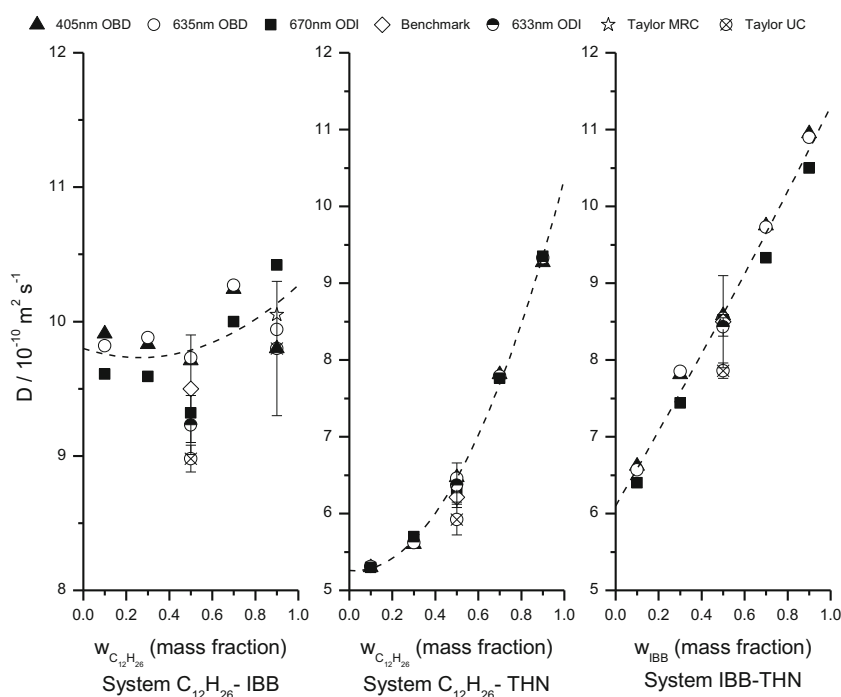
Results and Discussion

Tests of the Dispersion for Binary and Ternary Systems

The three possible binary mixtures (THN-IBB, IBB- $\text{C}_{12}\text{H}_{26}$ and THN- $\text{C}_{12}\text{H}_{26}$) at the 0.5:0.5 mass fraction symmetric points, and a fourth mixture THN- $\text{C}_{12}\text{H}_{26}$ with 0.9:0.1 mass fraction, were used to test the operation of the dispersion equipment. These systems were chosen because their diffusion coefficients are accurately known from other experimental methods, as of Gebhardt et al. (2013) and Platten et al. (2003); the deviations between the results obtained from these techniques are, in general, less than 7 %, and at the most 8.5 %. The point 0.9:0.1 for THN- $\text{C}_{12}\text{H}_{26}$ was also measured using the Taylor dispersion technique by Gebhardt et al. (2013) and provides possibilities for direct comparison. Taking these measurements as references, we have been able to adapt, optimize and build experimental protocol for our Taylor dispersion equipment which can be applied in the measurement of diffusion coefficients in the Fontainebleau benchmark mixture.

Table 1 gives the mean D values for four binary systems at 298.15 K determined from four to six replicate dispersion profiles. Comparative plots of our results with those previously published by Gebhardt et al. (2013), Platten et al. (2003) and Mialdun and Shevtsova (2011) for the same systems are shown on Fig. 1. Direct comparison of the results for the Taylor D values reported here,

Fig. 1 Binary diffusion coefficients for the Systems IBB/THN, $C_{12}H_{26}$ /IBB and $C_{12}H_{26}$ /THN



with D values obtained by other experimental methods, reveals that they are encompassed within the previously found 7 % uncertainty interval. If we consider that 1–3 % uncertainty is typical for Taylor dispersion measurements, as shown in previous reports for binary systems by Santos et al. (2012, 2013, 2015) and Ribeiro et al. (2006, 2010, 2011) this suggests an acceptable accuracy in these determinations.

The ternary diffusion coefficients measured for THN (1) + IBB (2) + $C_{12}H_{26}$ (0) system are summarized in Table 2. The component zero is for the solvent. The main diffusion coefficients D_{11} and D_{22} were normally reproducible within $\pm 0.08 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$. The cross-coefficients were, in general, reproducible within about $\pm 0.10 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$. The results obtained for the mixtures under study are in reasonable agreement with available results in the literature, measured using Taylor dispersion, Counter Flow Cell and Sliding Symmetric Tubes techniques by Mialdun et al. (2013), Sechenyh et al. (2013) and Larrañaga et al. (2013). The ternary results obtained here differ only slightly from the published ones, and, although there is still no agreement on the actual values, the eigenvalues obtained from all the experiments are very close. Our eigenvalues differ between 3.5 and 8.8 % from those measured with the same experimental method. For the symmetric point under study there is one published result by Koninger et al. (2010), which was obtained by the Optical Beam Deflection technique in this system with a different order of the components ($C_{12}H_{26}$ (1) + IBB (2) + THN (0)). Although this implies different values for the main and cross diffusion coefficients, the

eigenvalues of the diffusion matrix do not depend on the order of the components, and the differences between our and the OBD published values are 17.8 % in \widehat{D}_1 and 3.4 % in \widehat{D}_2 .

We can conclude from the non-zero cross diffusion coefficient values, D_{ij} , at this finite concentration for the THN (1) + IBB (2) + $C_{12}H_{26}$ (0) system, and by the fact that the main coefficients D_{ii} are not the same as the binary diffusion ones, that there are solute interactions affecting the diffusion of the components, and, under these conditions, possible interactions between the solutes leads to a counter flow of IBB ($D_{21} < 0$). The gradient in the concentration of IBB produces co-current coupled flows of THN. If we consider that D_{12}/D_{22} values give the number of moles of THN co-transported per mole of IBB driven by its own concentration gradient, we can suggest that a mole of diffusing IBB co-transport 0.14 mol of THN at the concentration studied. From the D_{21}/D_{11} values, at the same compositions, we can anticipate that a mole of diffusing THN counter-transport 0.44 mol of IBB.

Diffusion: Useful Strategy for Structural Interpretation of Chemical Systems

Limiting Diffusion Coefficients and Radii of the Diffusing Entities

From the analysis of our D experimental data, together with literature diffusion coefficients at infinitesimal concentration, D^0 by Gebhardt et al. (2013), we can obtain structural

information of the diffusing entities. The Stoke’s radii (r_s) have been estimated using the Stokes Einstein equation (5), which assumes that the particles are perfectly spherical and are subject solely to solvent friction (Erdey-Grúz 1974)

$$D^0 = k_B T / 6\pi \eta^0 r_s \tag{5}$$

where D^0 is the tracer diffusion coefficient of a species D^0 is the tracer diffusion coefficient of a species, and k_B and η^0 are Boltzmann’s constant, and the solvent viscosity at temperature T (see Aminabhavi and Gopalakrishna 1995, Rathnam et al. 2010). As can be seen from Table 3, the different structures of these compounds lead us to obtain significant differences between their D^0 values that do not necessarily translate into changes in their Stokes radii. For example, for dodecane ($C_{12}H_{26}$) there is only a slight difference in its radius when the solvent is IBB or THN. Since the only possible interactions between these three molecules are Van der Waals and London dispersion forces, the smaller value obtained for $C_{12}H_{26}$ in THN is probably related to steric constraints on the accommodation of these molecules in the liquid phase.

Estimation of the Activity Coefficients

The inexistence of available vapor-liquid equilibrium data for the binary and ternary systems under study, as far as the authors know, constrains the calculation of activity coefficients. Therefore an alternative model for estimation of activity coefficients is presented ahead.

For non-ideal and non-dilute system of two components, Fick’s first law describes the flux that occurs in the system as a result of a drive to approach thermodynamic equilibrium. This flux is caused by gradients of chemical potential of the components rather than concentration, and for one-dimensional, isothermal, isobaric diffusion with no external potential gradients comes described by

$$J_i = -D_i \frac{\partial c_i}{\partial z} = -M_{ii} \frac{\partial \mu_i}{\partial z} \tag{6}$$

where D is the diffusion coefficient, c is concentration, M_{ii} are the coupling coefficients between fluxes and reflect the driving force of the flux and the mobility of the species in responding with movement, μ_i is the chemical potential and z is the axis coordinate.

Introducing the definition of chemical potential for a given component

$$\mu_i = \mu_0(T, P) + k_B T \ln a_i = \mu_0 + k_B T (\ln x_i + \ln \gamma_i) \tag{7}$$

Table 3 Estimation of radii of three solutes ($C_{12}H_{26}$, IBB, THN) in different binary mixtures from their limiting binary diffusion coefficients, D^0 at 298.15 K

System	$D^0/10^{-9} \text{ m}^2 \text{ s}^{-1a}$	$R_s/10^{-9} \text{ m}$
$C_{12}H_{26}$ (1)/IBB (2)	0.980	0.231
$C_{12}H_{26}$ (1)/THN (2)	0.526	0.207
IBB (1)/THN (2)	0.610	0.179

^aGebhardt et al. (2013)

into Fick’s first law the expression for the flux is then:

$$J_i = -D_i \frac{\partial c_i}{\partial z} = -\frac{M_{ii} k_B T}{c_i} \left(1 + \frac{\partial \ln \gamma_i}{\partial \ln x_i} \right) \frac{\partial c_i}{\partial z} \tag{8}$$

where a_i is the activity of component i , γ_i is the activity coefficient of i , x_i is the mole fraction of component i k_B is Boltzmann’s constant and T stands for temperature.

If tracer diffusion coefficient that comes defined by Eq. 4, is the diffusion coefficient for ideal mixtures (where γ_i is constant), rearranging Eq. 7 we have

$$D_i = D_i^o \left(1 + \frac{\partial \ln \gamma_i}{\partial \ln x_i} \right) \tag{9}$$

where the parcel between brackets in normally called thermodynamic factor. By combining Eq. 8 with the one-parameter Margules (1895) activity model for a binary mixture of component 1 and component 2

$$\ln \gamma_1 = Ax_2^2 \quad \text{and} \quad \ln \gamma_2 = Ax_1^2 \tag{10}$$

activity coefficients can then be estimated from the molar fraction dependence of the ratio D/D^0 , considering through this approximation that variation in D is due to the variation of this thermodynamic factor.

The values thus obtained for the activity coefficients, γ , for the 3 different mixtures studied here are shown in Table 4. Data in Table 4 was obtained using molar fraction of the solutes in Eqs. 6–7; mass fraction information is present only for easier correlation with all other data. From the observed γ values for the system IBB/THN, we can say that in this particular mixture their components interact attractively toward each other and, consequently, this system is the one closest to the ideal solution, in contrast to what is observed for the other systems. That is, for $C_{12}H_{26}$ / IBB and $C_{12}H_{26}$ /THN systems, the γ values reflect mixtures that are further away from ideality, mainly for extreme molar fractions of the components. This is an expected behaviour taking into account the steric constrains found when mixing

Table 4 Solute activity coefficients, γ , for different binary mixtures of THN, IBB and C₁₂H₂₆ at 298.15 K

Binary System	w_1	γ_1	γ_2
C ₁₂ H ₂₆ (1)/IBB (2)	0.1	1.149	1.001
	0.3	1.096	1.009
	0.5	1.053	1.029
	0.7	1.021	1.064
	0.9	1.003	1.120
C ₁₂ H ₂₆ (1)/THN (2)	0.1	2.120	0.997
	0.3	1.647	0.973
	0.5	1.324	0.920
	0.7	1.119	0.835
	0.9	1.014	0.717
IBB(1)/THN (2)	0.1	0.998	1.000
	0.3	0.993	0.999
	0.5	0.996	0.998
	0.7	0.999	0.996
	0.9	1.000	0.993

two liquids composed with different shaped molecules, since it requires the rupture in the structure of the media.

Conclusions

Taylor dispersion equipment installed at the University of Coimbra for the measurement of diffusion in liquids has been tested to ensure in general an acceptable and adequate accuracy (2–5 %) by measuring mutual diffusion coefficients for binary aqueous solutions of systems.

Binary mutual diffusion coefficients for mixtures of THN, IBB and C₁₂H₂₆ at 0.5:0.5 mass fraction symmetric points, and for 0.9:0.1 mass fraction in IBB- C₁₂H₂₆ were measured with the Taylor dispersion technique. The results are in agreement with literature values and provide reliable support for data on this benchmark mixture.

Based on these measurements, it was possible, using appropriate models, to estimate transport, structural and thermodynamic parameters (D^0 , radius and activity coefficients), which help us understand the nature and structure of these mixtures, which are important for practical applications in various fields, including the oil industry.

The ternary system containing equal mass fraction of all the components (0.33 THN- 0.33 IBB –0.33 C₁₂H₂₆) was also studied at 298.15 K. Good agreement was observed for both the diffusion coefficients and respective eigenvalues.

Our Taylor dispersion equipment has been successfully optimized and we were able to positively contribute with consistent isothermal diffusion data to characterization of the Fontainebleau benchmark mixture.

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