DETERMINATION OF THE TRANSPORT COEFFICIENTS OF THE DCMIX1 TERNARY MIXTURES

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Introduction:
Transport properties in multicomponent mixtures have awakened a high level of interest in the scientific community, due to their involvement in different areas such as the oil industry, biology or engineering [1-4]. In binary mixtures, significant progress has been made in the understanding of the thermodiffusion phenomenon within the last decade. There are several experimental techniques to reliably determine transport coefficients [5-7]. However, mixtures with scientific or technological interest are generally multicomponent. In recent years, several works analysing ternary mixtures have been published [8-14] with the aim of achieving a better understanding of the thermodiffusion phenomenon in ternary mixtures. Recently, the first Benchmark in ternary mixtures [15] has been established, which has come as a great step forward. This has been developed thanks to the international collaboration project DCMIX (Diffusion coefficient Benchmark, ternary mixtures). This Benchmark incorporates the results of six independent teams utilising different experimental techniques and both in microgravity and ground conditions. Nevertheless, a wide data base of reliable experimental results is still necessary. This data base may be helpful for validating the theory that is still not unified [16-18]. Moreover, it is valid for developing numerical models or correlations that enable the prediction of the thermodiffusion coefficients [19-23].

The objective of this work is to contribute to the experimental data base in multicomponent mixtures. For that purpose, new experimental results for four ternary mixtures are provided. These mixtures correspond to the mixtures of the project DCMIX, which are formed by 1,2,3,4-tetrahydronaphthaline (THN), isobutylbenzene (IBB) and dodecane (nC12) at six different mass concentrations. In all the cases, the analysis has been completed at 25°C. In this work, THN is considered as component 1, IBB as component 2 and nC12 as component 3 in the mixture.

Material and methodology
The ternary mixtures analysed in this work are all composed of THN, IBB and nC12, with purity higher than 99%. Table 1 shows the compositions that have been studied.

<table>
<thead>
<tr>
<th>c (THN)</th>
<th>c (IBB)</th>
<th>c (nC12)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.10</td>
<td>0.80</td>
<td>0.10</td>
</tr>
<tr>
<td>0.10</td>
<td>0.10</td>
<td>0.80</td>
</tr>
<tr>
<td>0.45</td>
<td>0.10</td>
<td>0.45</td>
</tr>
<tr>
<td>0.40</td>
<td>0.20</td>
<td>0.40</td>
</tr>
<tr>
<td>0.33</td>
<td>0.33</td>
<td>0.33</td>
</tr>
</tbody>
</table>

Before determining transport coefficients in a mixture, it is necessary to define some thermophysical properties of it. To determine the density we used an Anton Paar DMA 5000 vibrating quartz U-tube densimeter with an accuracy of 5x10⁻⁵ kg/m³. The density measurements also provide the thermal expansion coefficient. Coupled to the densimeter is an Anton Paar RXA 156 refractometer with accuracy of 2x10⁻⁵ RIU, which measures the refractive index. Finally, an Anton Paar AMVn microviscometer was used for the determination of the dynamic viscosity.

To analyse a ternary mixture it is also necessary to make a calibration that enables the determination of the concentrations of each component from the measurements of the density and the refractive index. This calibration consists of measuring the density and the refractive index of 25 mixtures around the concentration of study. From this data the calibration coefficients a, a', b, b', c and c' can be calculated, which are used to determine the concentration of each component in the mixture by the following equations:

\[ c_1 = \frac{c(a-a')-c(nD-a)}{b'c-bc} \]  \hspace{1cm} (1)

\[ c_2 = \frac{b(nD-a')-b'(p-a)}{bc-b'c} \]  \hspace{1cm} (2)

\[ c_3 = 1 - c_1 - c_2 \]  \hspace{1cm} (3)

where \( c_i \) and \( nD \) are the concentrations of components 1, 2 and 3 respectively, \( p \) is the density of the mixture and \( nD \) is the refractive index of the mixture.

a) Thermogravitational technique:
The thermogravitational technique [22, 23] has been used to determine the thermodiffusion coefficients in this work. In this work the new thermogravitational column which was presented and validated with the Benchmark ternary mixture [24] was used. The gap of the column is 1±5x10⁻³ mm wide and 980 mm high.

By this technique, we can measure the concentration gradient along the height of the column (Fig. 1) which, together with the thermophysical properties, enables the determination of the thermodiffusion coefficients by the following equation:

\[ D'_{T,i} = -\frac{L_x^4}{504} \frac{g \alpha c}{\mu} \frac{\partial c_i}{\partial x} \]  \hspace{1cm} (4)

where \( L_x \) is the gap width; \( g \) is gravity; \( \alpha \) is the thermal expansion coefficient; \( \mu \) is the dynamic viscosity; and \( \partial c_i/\partial x \) is the concentration gradient of component \( i \) in the height of the column.

b) Sliding Symmetric Tubes technique:
The Sliding Symmetric Tubes (SST) technique [8, 20] has been used to determine the molecular diffusion coefficients in this work. By this technique, we measure the variation of the concentration with time (Fig. 2) and from that variation we can determine the molecular diffusion coefficients by the following working equations:

\[
S_1 = \frac{2}{L} \left( \frac{\sqrt{A}}{a_1} + \frac{\sqrt{B}}{a_2} \right) \tag{5}
\]

\[
S_2 = \frac{2}{L} \left( \frac{1-\sqrt{D_{11}a_1^2}}{a_1} + \frac{1-\sqrt{D_{12}a_2^2}}{a_2} \right) \tag{6}
\]

where \(S_1\) and \(S_2\) are the slopes of components 1 and 2, \(A\) and \(B\) are the integration constants, \(a_1\) and \(a_2\) are proportional to the eigenvalues of the diffusion matrix, \(L\) is the length of the tubes and \(D_{ij}\) and \(D_{ij}\) are two of the molecular diffusion coefficients. \(A\), \(B\), \(a_1\) and \(a_2\) are functions of the four diffusion coefficients.

c) Determination of Soret coefficient:

Finally, the Soret coefficient is determined from the results obtained for the thermodiffusion and the molecular diffusion coefficients. They are related by the following equations [12]:

\[
S'_{T,1} = \frac{\dot{b}_{11} - \dot{b}_{22}}{\dot{b}_{12}} \quad S'_{T,2} = \frac{\dot{b}_{21} - \dot{b}_{11}}{\dot{b}_{22}} \tag{7}
\]

Results

We have measured the thermodiffusion, molecular diffusion and Soret coefficients for the five ternary mixtures of Table 1. In addition, the thermophysical properties of these mixtures have been also determined.

These results complete the results measured for the Benchmark ternary mixture [15].

Discussion and conclusions

We have measured the thermodiffusion, molecular diffusion and Soret coefficients for the five mixtures of Table 1. In addition, the thermophysical properties of these mixtures have been also determined.

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References: