



## Solutal natural convection flows in ternary mixtures



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### ABSTRACT

It is known that the cross diffusion terms generate four different types of solutions to the one-dimensional unsteady diffusion equations for ternary mixtures. The stability of the fluid column corresponding to these solutions can be classified depending on the sign of the first derivative of density with respect to the direction of the gravity vector (i.e.  $\partial\rho/\partial y$ ) and the sign of  $(\partial^2\rho/\partial y^2)/y$ , where  $y = 0$  is located at the center of the diffusion layer. The type of solution depends on the initial conditions and on the set of diffusion coefficients considered. One type of solution corresponds to a stable fluid column with  $(\partial\rho/\partial y < 0)$  and  $(\partial^2\rho/\partial y^2)/y > 0$ . Two types of solutions generate fluid layers with unstable density stratification  $(\partial\rho/\partial y > 0)$  and the fourth type shows a fluid layer with  $(\partial^2\rho/\partial y^2)/y < 0$ . We analyzed the unsteady diffusion processes in a ternary mixture under the conditions of experiments carried out to determine the four diffusion coefficients of the ternary system 1,2,3,4-tetrahydronaphthalene-isobutylbenzene-dodecane (THN-IBB-nC12). These measurements; performed in diffusion cells with an initially stable stratification within the cell, with the denser mixture at the bottom and the lighter at the top; are usually based on the validity of the one-dimensional unsteady diffusion mass transfer equations. The linear stability analysis for the onset of convection in the unstable layers with unstable density stratification  $(\partial\rho/\partial y > 0)$  indicates that the critical thickness of these layers depends on the Rayleigh numbers, on the diffusion coefficients and on the initial conditions. To illustrate the flow structures that can be generated in these unstable conditions, we performed numerical simulations for selected sets of diffusion coefficients at different Rayleigh numbers. The results of these simulations are in general agreement with the predictions of the linear stability analysis and indicate that, under specific conditions, the convective motions developed in the cell produce significant departures of the concentration distributions from the pure diffusion situation.

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## 1. Introduction

The experimental determination of the mass diffusion and thermodiffusion coefficients of the different chemical species in multi-component liquid systems is relevant in many scientific and technological applications related, for instance, with crystal growth or biological systems [1–4]. Additionally, mass diffusion and thermodiffusion have, due to the geothermal gradient, important implications in Earth sciences and, in particular, in petroleum industry. In this case, a more precise understanding of thermodiffusion phenomena can result in a more accurate modeling of crude oil reservoirs, which can lead to the reduction of the number of wells required and the cost of the initial prospecting [5–7].

The present study considers liquid mixtures at room temperature -isothermal conditions- and the particular ternary mixture of 1,2,3,4-tetrahydronaphthalene (THN), isobutylbenzene (IBB) and n-dodecane (nC12). This mixture is considered as a representative model of the different molecular families of the oil in natural reservoirs. Specifically, the naphthalenic, aromatic and aliphatic compounds are associated with the THN, IBB and nC12, respectively. Due to this similarity, this particular system has been extensively investigated in Earth laboratories and also in reduced gravity environments, as the FOTON M3 spacecraft [8] or the International Space Station (ISS). In this last case, the corresponding experimental campaign of the European Space Agency, named Diffusion and Soret Coefficients-Diffusion Coefficients in Mixtures (DSC-DCMIX1) was initiated six years ago and many interesting results are still appearing in the literature [9–11].

A common experimental strategy for the determination of the four molecular diffusion coefficients of this transparent ternary

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**Nomenclature**

$a$	wavenumber
$D$	diffusion coefficient
$g$	gravitational acceleration
$K_e$	kinetic energy
$L$	characteristic length
$p$	pressure
$\Re$	real part
$Ra$	Rayleigh number
$Sc$	Schmidt number
$t$	time
$u_i$	velocity components
$w$	mass fraction
$x, y, z$	Cartesian coordinates

**Greek letters**

$\beta$	solubility expansion coefficient, $\beta_i = (\partial\rho/\partial w_i)/\rho_{(0,0)}$
$\delta_{ij}$	Kronecker's delta
$\delta$	layer thickness
$\Delta$	increment
$\eta$	similarity variable
$\lambda$	wavelength

$\mu$	dynamic viscosity
$\nu$	kinematic viscosity
$\rho$	density
$\psi$	streamfunction

**Subscripts and superscripts**

*	non-dimensional
1, 2, 3	component of the mixture
$b$	basic state
$bot$	value at the bottom of the cell
$c$	critical
$in$	initial value
$o$	reference value
$top$	value at the top of the cell

**Special characters**

$\bar{\phantom{x}}$	averaged on a horizontal plane
$\langle \phantom{x} \rangle$	volume averaged
$'$	perturbation

system on Earth laboratories and at room temperature consists in the initial introduction of a heavier mixture at the bottom part of a diffusion cell while a lighter one is placed at the top. In this situation of stable stratification, molecular diffusion takes place and the resulting concentration distribution allows the final obtaining of the different terms of the diffusion matrix. In the Counter Flow Cell (CFC) technique [12,13] optical digital interferometry [14–16] is used to measure indirectly the density profile within the mixture. The subsequent nonlinear fitting between the obtained experimental data and the corresponding theoretical profiles of the purely diffusive process allows the obtaining of the four diffusion coefficients [12,13]. Another methodology, the so-called Sliding Symmetric Tubes (SST) technique, uses a set of large moving tubes of small diameter [17–19]. The measurements and the subsequent nonlinear regressions are based on the determination of the averaged concentration values of the two different species in the upper and lower part of the different tubes [20,21]. The typical length of the parallelepipedic cell in the CFC method is about one centimeter while that of the cylindrical cell in the SST one is fifty centimeters. This means that the experiments, especially in the second case, are very time consuming. Typically, fifteen–eighteen days for SST against a few hours for CFC. However, despite the apparent simplicity of both methods, there exists in the literature an evident scatter in the numerical values of the diffusion coefficients which indicates some still unsolved difficulties [20].

To help in the resolution of this problem other methodologies have been used in Earth laboratories, not only to obtain the diffusion matrix but also the Soret and thermodiffusion coefficients. The most common techniques found in the literature are the Open Ended Capillary Technique (OCT) [22,23], the Taylor Dispersion Technique (TDT) [24] and the Two-Color Optical Beam Deflection Technique, (OBD) [25].

In addition, to try to avoid the potential effect of residual convection in diffusion cells, the DSC-DCMIX1 experiment has also been performed in the International Space Station. In this case, the determination of the different coefficients was performed using the Selectable Optical Diagnostic Instrument (SODI) installed in the Microgravity Science Glovebox on the U.S. Laboratory, Destiny module [26]. The application of these different measure-

ment techniques and environments to the same mixture shows appreciable scatter of the results, mainly in the values of the secondary diagonal of the diffusion matrix.

In Earth laboratories, a potential source of problems could be the evolution of convective motions in the initially stable flow due to the local competitions of the two driving forces generated by the concentration gradients of both independent components [27–29]. The result of this competition generates, in some cases, double-diffusive instabilities which could be appreciable perturbations in the experimental determination of the four molecular diffusion coefficients [30–33].

In summary, the present study particularizes and analyzes, theoretically and numerically, the possibility of the onset of undesired solubility natural convection flows in diffusion experiments of the ternary mixture THN-IBB-nC<sub>12</sub> used in the DSC-DCMIX1 experiment. The flow structures that may develop and their potential effect on the determination of the diffusion coefficients are also reported and in deep discussed.

**2. Modeling details**

The THN-IBB-nC<sub>12</sub> ternary mixture at constant room temperature is enclosed in a small parallelepipedic cell with rigid walls. Fig. 1 shows the geometry used, which coincides with the one reported in the literature to determine the mass diffusion coefficients of the above-mentioned ternary mixture [12,13].

The density of the mixture as a function of the concentrations can be modeled as, [17,19]

$$\rho = \rho_0[1 + \beta_1(w_1 - \langle w_1 \rangle) + \beta_2(w_2 - \langle w_2 \rangle)] \quad (1)$$

where  $w_1$  and  $w_2$  are the local mass fractions of THN and IBB, respectively,  $\langle w_1 \rangle$  and  $\langle w_2 \rangle$  are the volume averaged mass fractions,  $\rho_0 = 842.377 \text{ kg m}^{-3}$ ,  $\beta_1 = 0.2581$  and  $\beta_2 = 0.1370$ . These values are valid for mass fractions ratios close to 1:1:1. The dynamic viscosity of the mixture is considered constant ( $\mu = 1.289 \cdot 10^{-3} \text{ Pa}\cdot\text{s}$ ) [17].

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