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Validity limits of the FJO thermogravitational column theory: Experimental and numerical analysis

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ABSTRACT

In this article, the limits of the validity of the Furry, Jones and Onsager theory (FJO) has been studied both numerically and experimentally. The commercial Solver Fluent® (Fluent Inc.) has been used in this study, as a computational fluid dynamics (CFD) tool. It was used in order to simulate the variations of the steady separation that will take place in a thermogravitational column, over a wide range of the product of the Grashof number (*Gr*) and the Schmidt number (*Sc*). On the other hand, we have used a thermogravitational column for experimental measurements of the achieved stationary separation in two dilute mixtures of polystyrene in toluene. Separation measures were taken at different values of Grashof and Schmidt numbers.

Both the numerical and experimental results show the validity of the FJO theory whenever the work conditions satisfy the relation: $Gr\,Sc \leqslant 1000A$, were A is the aspect ratio between the height of the thermogravitational column and the work gap (annular space). Outside the validity range the separation strongly decreases with respect to that given by the FJO theory.

1. Introduction

The theoretical description of the thermogravitational column was first developed by Furry, Jones and Onsager (FJO) [1]. These authors, introducing some simplifying assumption in the hydrodynamic equations, arrived to a basic relation that allows one to determinate the thermal diffusion coefficient of a liquid mixture from steady separation measurements [2]. In order to validate these approximations we have numerically solved the full thermohydrodynamic equations in 2D comparing the results obtained with the ones given by the FJO theory [3]. Using the numerical results obtained for the parallel plane approximation, we have shown in [3] that the validity range of this FJO theory must satisfy the following condition represented in Eq. (1):

$$Gr\,Sc < 1000A \tag{1}$$

with A the aspect ratio, Gr the Grashof number and Sc the Schmidt number given by: $A = L/d$, $Gr = g\alpha d^3 \Delta T/\nu^2$ and $Sc = \nu/D$, where α, is the thermal expansion coefficient, ΔT, the temperature difference between the column walls, d, the gap width, ν the kinematic viscosity, L the length of the column, g is the gravitational field and D the ordinary diffusion coefficient.

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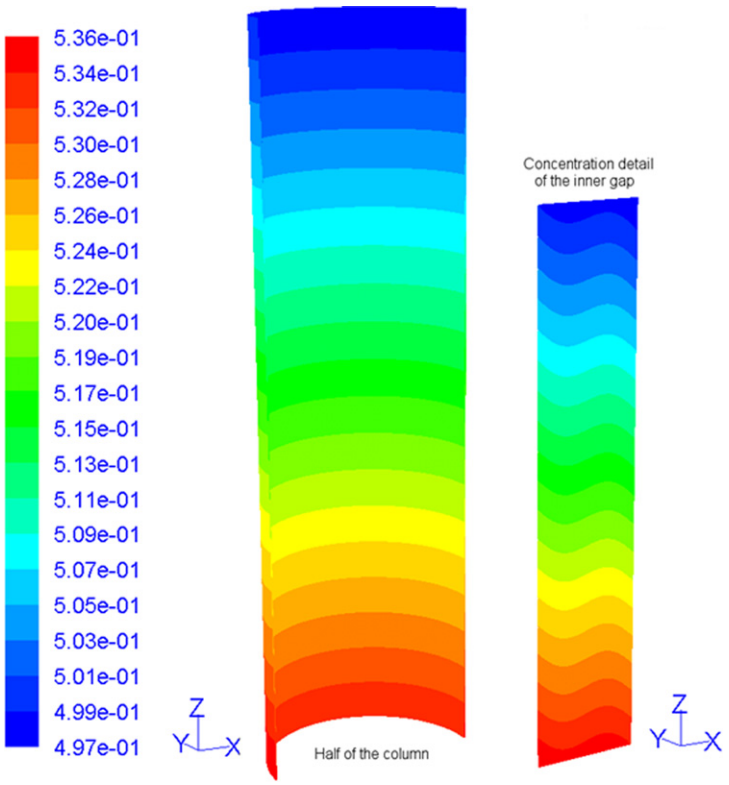

Fig. 1. Concentration of toluene (stationary state) in a thermogravitational column for the mixture of toluene/*n*-hexane. (Mass fraction of toluene of $c = 51.67\%$ and at average temperature of 298.16 K.)

In this study, first of all and in order to verify the results obtained in [3], the numerical resolution of the thermohydrodynamics equations was carried out for a cylindrical geometry in 3D based on a model of computational fluid dynamics (CFD) using the commercial solver of finite volumes Ansys® Fluent® 12.1. In second place, we have performed separation measurements in a thermogravitational column for different values of *Gr* and *Sc* in order to determine experimentally the reliability of the above condition. Two dilute mixtures of polystyrene in toluene have been employed.

2. Numerical analysis

The geometry of the thermogravitational columns allows one to work as much in 2D as in 3D, due to its symmetry with respect to the three axes. These symmetries allow us, to simplify the generated mesh in Ansys® Design Model® and also, to reduce the calculation time.

The results obtained by H. Barrutia et al. [4] using the finite volume method Ansys® Fluent® 6.3 were validated for the case of the natural convection, confined in both the parallelepiped and cylindrical configuration. The results showed the potential of this tool to determine the validity of the approach of the conductive regime. In this case, Ansys® Fluent® 12.1 was used to simulate the thermogravitational effect in a liquid binary mixture, that is, the convective behavior associated with the diffusive one.

A mesh was created in the pre-processor Ansys® Gambit® with the aim of discretizing the governing equations for fluid flow. This mesh is composed of 80 000 hexahedral cells, forming a coaxial cylinder.

A variable height has been considered in order to work with different aspect ratios, whereas the radial difference was maintained constant. The computational domain was discretized with structured hexahedral meshes, maintaining the value of aspect ratio between 0.1 and 0.2.

To simulate the thermogravitational behavior using Ansys Fluent®, the species transport model was activated along with the thermal diffusion option. The corresponding thermophysical properties of the mixture as well as those of the pure components were introduced. Then, the three equations of flow, momentum conservation, energy, and species were solved. The convergence criteria were lowered to 10^{-8}. In the numerical simulation, the variation of density as a function of temperature and concentration of the mixture was considered, including a linear function equation (2) by means of a user defined function (UDF):

$$\rho = \rho_0 \left[1 - \alpha \frac{\partial \rho}{\partial T} + \beta \frac{\partial \rho}{\partial c} \right] \tag{2}$$

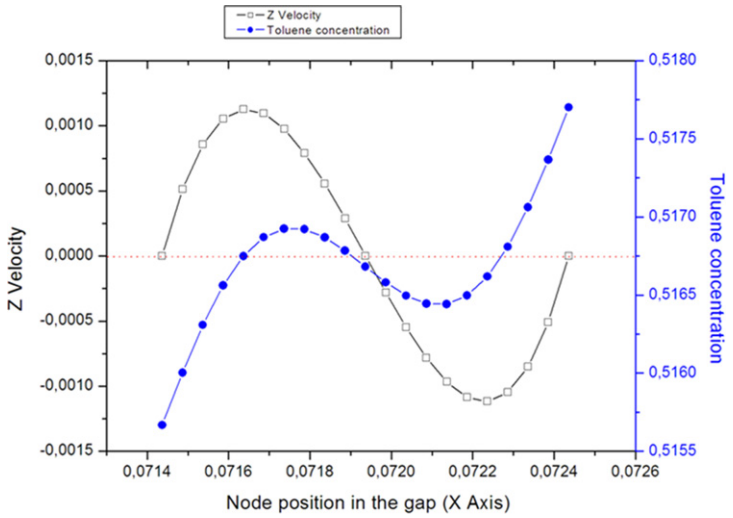

Fig. 2. The velocity and the toluene concentration profile inside the gap of the thermogravitational column to a given height of 0.25 m. (Toluene/*n*-hexane mixture with mass fraction of toluene of $c = 51.67\%$, and an average temperature of 298.16 K.)

Table 1
Separation of mass fraction obtained experimentally Δc (exp.) and from numerical analysis Δc (Fluent) for toluene–*n*-hexane (Tol-Hex) ($c_0 = 51.67$ wt% toluene) and mixtures of the benchmark Fontainebleau (Ref. [6]).

Mixture	Δc (Fluent)	Δc (exp.)	Error %
Tol-Hex	0.0360	0.0367	1.9
THN-IBB	0.0308	0.0313	1.6
THN-C12	0.0791	0.0785	−0.8
IBB-C12	0.0365	0.0376	2.9

where $\alpha = \frac{1}{\rho}\frac{\partial \rho}{\partial T}$ and $\beta = \frac{1}{\rho}\frac{\partial \rho}{\partial c}$, are, respectively, the thermal expansion coefficient and the mass expansion coefficient referring to the densest component of the mixture and of mass concentration c.

For the toluene–*n*-hexane ($c_0 = 51.67$ wt% toluene) mixture, Fig. 1 shows the distribution of the concentration referring to the densest component (toluene) in the stationary state. The results show an increase in the concentration of the densest component (toluene) in the lower and *n*-hexane in the upper parts of the column.

Fig. 2 shows the velocity and the toluene concentration profiles inside the gap to a given thermogravitational column height of 0.25 m. The velocity has a cubical profile while the concentration has a fifth order one. Column dimensions used in the calculations were: height of 50 cm and a gap of 1 mm. The thermophysical properties used for this mixture were taken from Refs. [5] and [6].

The results calculated for the separation were compared with the obtained experimental values in the laboratory using a thermogravitational column with an aspect ratio value of 500 [5]. These values are shown in Table 1, which indicate that the error between the experimental stationary values and the ones calculated by Fluent® is roughly 2%. These verifications were carried out for different liquid mixtures with confirmed thermophysical properties, for example the mixtures corresponding to benchmark of Fontainebleau [7,8]: (1,2,3,4-tetrahydronaftalene–isobutylbencene (THN-IBB), 1,2,3,4-tetrahydronaftalene–*n* dodecane (THN-C12), isobutylbencene–*n* dodecane (IBB-C12).

The experimental values obtained in a column with an aspect ratio value of 500 [5] and calculated separations inside the column are shown in Table 1. The deviations found between the numerical and the experimental separation is lower than 3%, within the considered experimental error. Therefore Fluent® was validated to provide and predict the separation values that would take place between the ends parts of the thermal diffusion column.

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3. Experimental method

The thermogravitational column used in this work is a conventional stainless-steel concentric tube type closed at both ends. It has been described in detail in earlier publications; for example, see Ref. [9] and references therein. The total length of the column is $L = 49$ cm and the distance between the sampling ports is $\Delta z = 40$ cm. The annular gap width is $d = 0.1$ cm being thus the aspect ratio $A = 490$ ($A = L/d$).

Table 2
Thermophysical properties of the polystyrene–toluene mixtures: kinematic viscosity (ν), ordinary diffusion coefficient (D), thermal diffusion coefficient (D_T). $(\Delta c)_{\mathrm{FJO}}$ is the separation in the FJO theory, Gr, Grashof number, Sc, Schmidt number, M is the mass of polystyrene and c_0 is the initial concentration.

M	c_0 (kg/m^3)	ν 10^{-6} (m^2/s)	D 10^{-11} (m^2/s)	D_T 10^{-11} (m^2/sK)	$(\Delta c)_{\mathrm{FJO}}$ 10^2	Gr ($\Delta T = 5$ K)	Sc
45 000	20	1.00	9.83	1.10	0.42	52.9	10 172
300 000	10	1.33	3.76	1.10	0.27	29.9	35 372

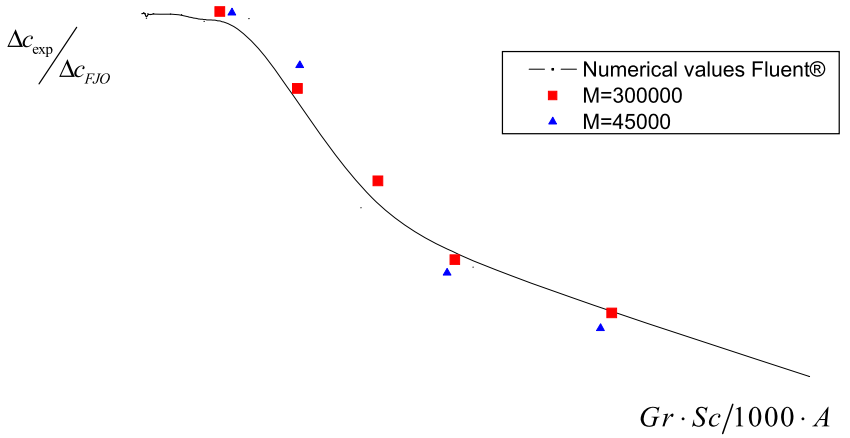

Fig. 3. Separation values compared to that obtained by FJO theory, $\Delta c/(\Delta c)_{\mathrm{FJO}}$ depending on $GrSc/1000A$; ■: $M = 300\,000$; ▲: $M = 45\,000$; _____: numerical results.

In order to have large enough values of the product $GrSc$ we have worked with polystyrene–toluene mixtures in the dilute region. Polystyrene samples of two molecular masses $M = 45\,000$ and $M = 300\,000$ at the concentration of 10 g/l and 20 g/l were used, respectively. The Schmidt number of these mixtures is of the order of 10^4.

The values of the diffusion coefficient, D, and the thermal diffusion coefficient, D_T, of the mixtures have been taken from the literature [10,11] and are shown in Table 2. The kinematic viscosity, ν, was measured in the Public University of Navarra using an Ubbelhode viscosimeter and it appears in Table 2. We have used the thermal expansion coefficient of the pure toluene ($\alpha = 1.08 \times 10^{-3}$ K^{-1}) for the thermal expansion coefficient of the dilute mixtures.

Separations, $(\Delta c)_{\mathrm{exp}}$, were determined by measuring the concentrations in the top and the bottom of the column with a Pulfrich refractometer with a nominal accuracy of 5×10^{-6} nD.

4. Results and discussion

We have determined the steady separation from FJO standard equation, represented in Eq. (3), using the physical properties in Table 2

$$\Delta c = c_0(1 - c_0)\Psi\left(\frac{D_T \nu}{\alpha}\right) \tag{3}$$

where Δc, is the separation; c_0, the initial mass fraction of polystyrene and a geometrical constant give by $\Psi = 504\Delta z/gd^4$, being Δz the distance between the sampling ports. The values obtained appear in Table 2 as $(\Delta c)_{\mathrm{FJO}}$. The values of the Grashof for $\Delta T = 5$ K and the Schmidt numbers for the considered mixtures are shown in Table 2. Temperatures differences between 3 K and 20 K have been used in order to operate with different values of the Grashof number.

In Fig. 3 are plotted the values of $(\Delta c)_{\mathrm{exp}}/(\Delta c)_{\mathrm{FJO}}$ as a function of $GrSc/1000A$. As it can be seen when this ratio is about unity the measured values of the separation Δc coincide with $(\Delta c)_{\mathrm{FJO}}$, indicating that the thermogravitational method can be used to determine the thermal diffusion coefficient of a polymer mixture when the column operates inside the validity range of the FJO theory that is when $GrSc \leqslant 1000A$. On the contrary, the measured separations for higher values of this ratio according to the numerical results, are smaller than the ones given by the FJO theory.

In Fig. 3 also are shown the numerical results obtained for this mixture of polystyrene–toluene in different $GrSc$ products. The results confirm the previous conclusions.

5. Conclusion

In this study, measurements of stationary separation in a thermogravitational column in dilute mixtures of polystyrene in toluene have been carried out. From this measures and the numerical results we can conclude that to determine reliable values of the thermal diffusion coefficient for liquid mixtures, the limits of validity of the FJO theory must be kept in mind

($Gr\,Sc \leqslant 1000A$). In turn, consequently the CFD Ansys® Fluent® commercial software is valid to predict the liquid binary mixtures thermohydrodynamic behavior under the thermogravitational effect.

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