Separation stability in binary mixtures with negative Soret numbers

B. ŠETA[1], E. LAPEIRA[2], D. DUBERT[1], F. GAVALDA[1], M.M. BOU-ALI[2], X. RUIZ[1]

1 – Department of Physical and Inorganic Chemistry, Universitat Rovira i Virgili,
C/ marcel.li Domingo s/n, 43007 Tarragona, Spain
2 - Mechanical and Industrial Manufacturing Department, MGEP Mondragon Goi Eskola Politeknikoa,
Loramendi 4 Apdo. 23, 20500 Mondragon, Spain
berin.seta@urv.cat

Abstract

In the present work, by using a paralelepipedic thermogravitational microcolumn, the temperature gradient influence on the stability of the flow was or has been examined, emphasizing mixtures with negative Soret coefficients. Experiments and numerical analysis were conducted for DCMIX2 Toulene-Methanol binary subsystem. This binary subsystem has a broad range of negative Soret values for low concentrations of Methanol which was analysed. Two different concentrations have been studied in order to confirm existence of temporal stability windows of those mixtures. Experiments were compared with numerical simulations conducted in open source software OpenFOAM, for both cases.

Keywords: *negative Soret, separation stability, binary mixtures, themogravitational microcolumn*

1. Introduction

Thermal diffusion phenomenon plays an important role in many technological and biological separation processes [1,2,3]. Thermodiffusion experiments are constructed in order to obtain Soret coefficients, which is linked with therrmodiffusion coefficients obtained by thermogravitational technique [1] and molecular diffusion coefficients obtained by pure diffusion experiments like those in Sliding Symmetric tubes, Open Ended Capillary tube or Taylor dispersion technique [4,5,6]. In our case, liquid binary mixture is placed between two parallel closed walls maintained on different temperature levels. Due to this gradient, the Soret effect appears and horizontal separation is provoked. If the Soret coefficient of the mixture is positive for denser component, denser component goes to cold wall and less dense component migrates toward hot wall. Due to natural convection, less dense component rises to the upper part of column, while denser goes to the bottom. For negative Soret coefficients, behavior will be opposite, hence creating unstable stratification of density inside column. Sketch of thermogravitational column can be found on figure 1.

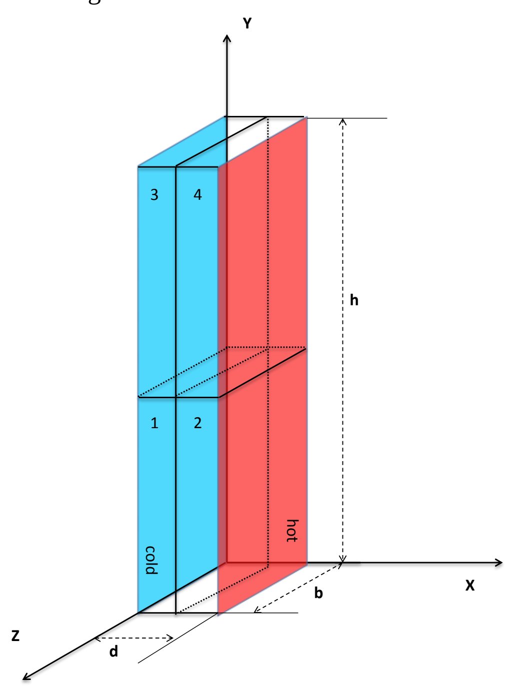

Fig. 1. Sketch of the thermogravitational microcolumn

2. Experimental procedure

This section narrates the experimental procedure and techniques followed for the determination of thermophysical properties of the binary mixtures. It is important to mention that thermogravitational microcolumn has dimension of d=0.51mm, b=3mm and h=30mm, making volume of column sufficiently small to operate with expensive or rare mixture such are biological fluids. Optical digital interferometry is used in determination of transport properties inside fluid, in our case, thermodiffusion coefficient. Further information about complete experimental procedure can be found in [7].

2.1 Thermophysical properties

Work is conducted with two different concentrations of Toulene-Methanol binary mixture, for two different Soret negative numbers.

Table 1: Thermophysical properties of two different working concentrations of Toluene-Methanol mixture

Case/ Physical properties	Mixture 1	Mixture 2
c_0 (toluene)	0.908	0.842
$1-c_0$(methanol)	0.092	0.158
ρ_{mean} (kg/m^3)	854.772	849.597
ν (m^2/s)	6.376	6.615
β_T (*10^{-3}) (K^{-1})	1.113	1.117
β_c (*10^{-2})	9.09	9.09
α(*10^{-8}) (m^2/s)$^{-1}$	9.564	9.564
D (*10^{-9}m^2/s)	1.14	0.9
D_T(*10^{-12} m^2*K/s)	-8.2	-3.375
S_T(*10^{-3} K^{-1})	-7.19	-3.75

Where c is concentration, ν is kinematic viscosity, β_T is thermal expansion coefficient, β_c is mass expansion coefficient, D is molecular diffusion coefficient, D_T is themodiffusion coefficient and S_T is Soret coefficient. As it is possible to see from table 1, mixture 1 has higher negative Soret number.

2.2 Numerical model

The continuity, momentum and mass transfer equations, assuming constant thermophysical properties of the mixture except for the linear variation of density with the temperature and concentration in the buoyancy terms (Boussinesq hypothesis), can be written as [9]:

$$\frac{\partial u_i}{\partial x_i} = 0 \tag{1}$$

$$\frac{\partial u_i}{\partial t} + \frac{\partial u_j u_i}{\partial x_j} = -\frac{\partial p}{\partial x_i} + \mu \frac{\partial^2 u_i}{\partial x_j{}^2} + \rho_0 g\left[1 + \beta_c\left(c_0 - c_{0,ref}\right) - \beta_T\left(T - T_{ref}\right)\right] \tag{2}$$

$$\frac{\partial c_0}{\partial t} + \frac{\partial u_j c_0}{\partial x_j} = D\frac{\partial^2 c_0}{\partial x_j{}^2} + D_T\, c_0(1-c_0)\frac{\partial^2 T}{\partial x_j{}^2} \tag{3}$$

$$\frac{\partial T}{\partial t} + \frac{\partial u_j T}{\partial x_j} = \alpha \frac{\partial^2 T}{\partial x_j{}^2}. \tag{4}$$

Non-slip impermeable boundary conditions for velocity are imposed at the six walls of the domain,

$$u_i\left(x_i, t\right) = 0, \quad x_i \in \Omega_w, \quad \forall t \geq 0, \quad i = 1, 2, 3.$$

Regarding the concentration, zero mass flux through the adiabatic walls,

$$\frac{\partial w_j(x_i, t)}{\partial x_i} = 0, \quad x_i \in \Omega_{adia,i}, \quad \forall t \geq 0, \quad i = 1, 2, \quad j = 1,$$

is used as boundary condition. Non-homogeneous Neumann boundary condition is imposed on the tempered walls,

$$\frac{\partial c_j(x_i, t)}{\partial x_i} = -S_{Ti} c_0 (1 - c_0)\frac{\partial T(x_i, t)}{\partial x_i}, \quad x_i \in \Omega_{T,i}, \quad \forall t \geq 0, \quad i = 3, \quad j = 1$$

3. Results

Stability windows for both mixtures is obtained numerically. This is possible to observe on figure 2. Its clearly shown that for mixture 1, that length of stability window was different for different temperature gradient applied, where higher temperature gradient resulted in longer stability window. For temperature different of 5K, theorical value was not reached at all.

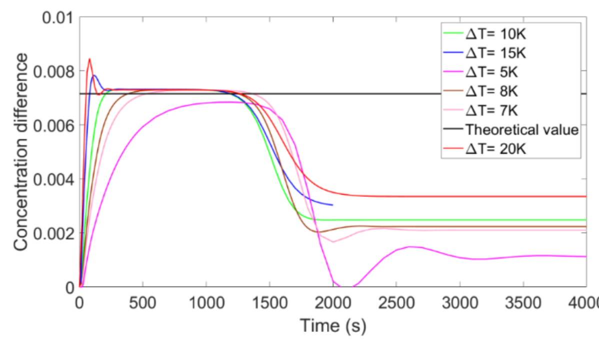

Figure 2. Separation stability windows for different initial temperature gradients.

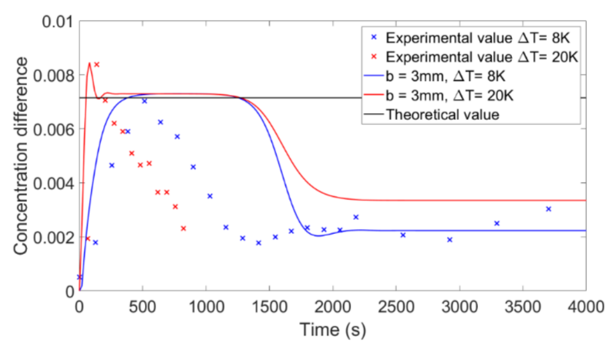

Fig. 3. Experimental and numerical separation in Tol(0.908)-Met(0.092).

Experimentally, mixture 1 was observed for $\Delta T = 8K \; and \; \Delta T = 20K$. Theoretical values were reached, but stability windows were not observed. This is very likely due to high negative Soret number. Behavior is shown on figure 3.

For mixture 2 and $\Delta T = 15K$, both theoretical value and stability window were observed experimentally. However, length of window inside in experiment was significantly shorter then in simulation. On figure 4, it is possible to compare transient separation in simulation and experiment for this case.

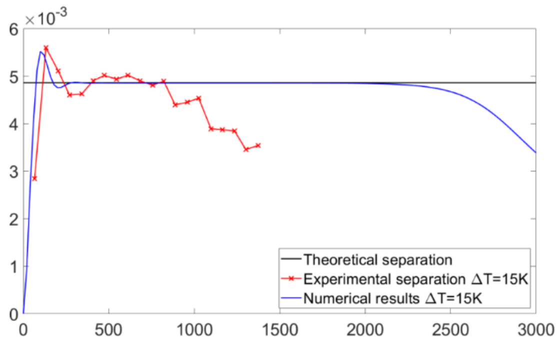

Fig. 4 Experimental run with ΔT=15 ºC in comparison with numerical results

4. Conclusions

This work brought new information about separation stability in binary mixture with negative Soret numbers. Stability windows are experimentally observed for first time in thermogravitational microcolumn, showing good coincidence with simulations.

Although, both theoretical value of separation and stability windows are reached in experiment, they are significantly shorter then in numerical simulations. This is probably due to imperfections in experimental setup, which are slightly different temperature gradient, orientation of thermogravitational column and slight differences in concentrations.

Future work will include ternary mixtures, where separation process will be much more complex.

Acknowledgments

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