



Thermodiffusion and coupled phenomena / Thermodiffusion et phénomènes couplés

## Thermodiffusion phenomena

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### ABSTRACT

The aim of this article is to present briefly a summary of the state of art in theoretical, experimental and numerical approaches in thermodiffusion. The concepts and equations giving the mass flux of constituents in binary, ternary and multicomponent mixtures are presented.

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## 1. Thermodiffusion

The physical phenomenon of thermodiffusion was initially discovered in the nineteenth century by Ludwig (1856) and by Soret (1879). It is a physical process occurring when a thermal gradient is applied to a mixture. Now it is known as the Ludwig–Soret effect (or more simply, the Soret effect).

The temperature field creates a concentration gradient in the mixture and, for a binary mixture, the mass flux is given by:

$$J_m = -\rho D \nabla C - \rho F(C) D_T \nabla T \quad (1)$$

where:  $\nabla C$  is the gradient of mass fraction induced by the temperature gradient  $\nabla T$ ,  $D$  is the coefficient of mass diffusion,  $D_T$  the coefficient of thermodiffusion,  $\rho$  the density,  $C$  the mass fraction of the considered component,  $F(C)$  is particular function of  $C$  verifying  $F(C=0)=0$  and  $F(C=1)=0$ . So, most of the authors use  $F(C)=C(1-C)$  and make the assumption that  $C(1-C) \approx C_0(1-C_0)$  where  $C_0$  is the initial value of the mass fraction. Let us note that this assumption supposes only small mass fraction variations around  $C_0$  and the validity of this assumption should be checked in each application. Moreover, with this simple mass fraction dependence of  $F(C)$ ,  $D_T$  remains mass fraction dependent and even may change sign with  $C$  as e.g. in water–ethanol systems, Kolodner et al. [1].  $D_T$  is also temperature dependent and may change sign with  $T$  as e.g. in water–NaCl solutions near 12 °C (Cadwell, in Mojtabi et al. [2]). Therefore treating  $D_T$  as a constant remains an additional assumption.

Under steady-state conditions  $J_m = 0$ , and with all the above mentioned assumptions, we have:

$$\nabla C = -\frac{D_T}{D} C_0(1-C_0) \nabla T \quad (2)$$

The ratio  $\frac{D_T}{D} = S_T$  is called the Soret coefficient.

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If  $\frac{D_T}{D} = S_T > 0$ , then  $\nabla C$  is of opposite sign to  $\nabla T$ . In other words,  $C$  decreases approaching the hot wall of the container. The absolute value of  $S_T$  remains rather small and usually does not exceed  $10^{-2} \text{ K}^{-1}$ . Therefore, the mass fraction gradient induced by the Soret effect remains small, but nevertheless in many important natural, physical or technological processes it has been shown that thermodiffusion phenomena plays a crucial role. Particular natural examples are: thermohaline convection in oceans driven by salinity gradients associated with temperature differences, mass transport across biological membranes induced by small thermal gradients in living matter where thermal diffusion could assume a sizable magnitude for changes of K–Na ionic activities within brain cells or an organ or a tumor.

Thermal diffusion has various applications, such as isotope separation in liquid and gaseous mixtures, characterization and separation of polymers, surface coating, etc. This phenomenon is supposed to play an important role in crystal growth process.

Soret-driven convective effects cannot be neglected in many industrial processes. Thermal diffusion, together with isothermal and pressure diffusion, may significantly affect the initial state of petroleum reservoirs.

## 2. Thermogravitational diffusion in binary fluid

The combination of two phenomena, convection and thermodiffusion, is called thermogravitational diffusion. The coupling of these two phenomena leads, in some circumstances, to large species separation.

In 1938, Clusius and Dickel [3] successfully carried out the separation of gas mixtures in a vertical cavity heated from the side (thermogravitational column, TGC). During the following years, two fundamental and experimental works on species separation in binary mixtures by the thermogravitational effect were published. Furry et al. [4] developed the theory to interpret the experimental processes of isotope separation (FJO theory). Subsequently, many works appeared, aimed at justifying the assumptions or extending the results of the FJO theory as, for example, to the case of binary liquids [4].

The fundamental parameter characterizing the influence of thermal diffusion on the convection in a binary mixture is the separation ratio  $\psi$ , see Eq. (7). For the binary mixture the density is generally function of the temperature and the mass fraction:  $\rho = \rho(T, C)$ , we deduce that the density gradient is function of the temperature and the mass fraction gradient:

$$\nabla \rho = \left( \frac{\partial \rho}{\partial T} \right)_C \nabla T + \left( \frac{\partial \rho}{\partial C} \right)_T \nabla C \quad (3)$$

For a small variation of  $T$  and  $C$ , the density varies linearly with local temperature and mass fraction:

$$\rho = \rho_0 (1 - \beta_T (T - T_0) - \beta_C (C - C_0)) \quad (4)$$

where  $\beta_T$  and  $\beta_C$  are the thermal and solutal expansion coefficients with:

$$\beta_T = - \left( \frac{1}{\rho_0} \frac{\partial \rho}{\partial T} \right)_{T_0, C_0} \quad \text{and} \quad \beta_C = - \left( \frac{1}{\rho_0} \frac{\partial \rho}{\partial C} \right)_{T_0, C_0} \quad (5)$$

Note that  $\beta_C$  is positive (negative) if  $C$  is the mass fraction of the lighter (heavier) component.  $\beta_T$  is generally positive. For the mechanical equilibrium steady state, the gradient of mass fraction  $\nabla C$  is given by the relation (2). Replacing  $\nabla C$  in the relation (3) we obtain:

$$\nabla \rho = -\rho_0 \beta_T \left( 1 - \frac{\beta_C}{\beta_T} \frac{D_T}{D} C_0 (1 - C_0) \right) \nabla T = -\rho_0 \beta_T (1 + \psi) \nabla T \quad (6)$$

The separation ratio can be also expressed in terms of the Soret coefficient by:

$$\psi = - \frac{\beta_C}{\beta_T} C_0 (1 - C_0) \frac{D_T}{D} = - \frac{\beta_C}{\beta_T} C_0 (1 - C_0) S_T \quad (7)$$

Let us note that the sign of  $\psi$  is independent of the choice of the reference component: indeed, if we skip from the denser to the lighter component for the reference,  $D_T$  as well as  $\beta_C$  will change sign leaving the sign of  $\psi$  unchanged.  $\psi$  represents the mass fraction contribution to the buoyancy force relative the thermal contribution, independent of any choice of the reference component.

In the presence of gravity, the density gradient (versus the spatial orientation) can cause convective motion. In this case, the separation ratio characterizes the influence of thermal diffusion on the buoyancy force. We deduce from Eq. (6) that with  $\psi > 0$  the variation of the density is more pronounced when thermodiffusion is taken into account, rather than the variation of  $\rho$  caused by thermal expansion alone. For the separation ratio in the range  $-1 < \psi < 0$  the variation of the density is smaller. We can affirm that for  $\psi > 0$ , the linear critical Rayleigh number associated to the onset of convection in the Rayleigh–Bénard problem with binary fluid is smaller than the classical one associated with mono-constituent fluid in an infinite horizontal cell ( $Rac = 1707.76$ ) and bigger for  $-1 < \psi < 0$ . This dimensionless parameter is widely used in describing convection in binary systems with the Soret effect [5–14].

Other works were related to the improvement of experimental devices to increase separation. One way to adjust the velocity and to increase the separation is to tilt the cell by a given angle from the vertical. In this case, the horizontal

component of the temperature gradient decreases, which decreases the buoyancy force and the velocity of the flow. The tilted cell has already been used by De Groot [5] in the case of bulk fluid but for different forms of the cells. Platten et al. [15] used an inclined cavity, keeping the hot plate on the top, to increase separation. Elhajjar et al. [16] used a horizontal cavity heated from above with temperature gradients imposed on the horizontal walls to improve the separation process with the use of two control parameters. Lorenz and Emery [17] proposed the introduction of a porous medium into the cavity.

### 3. Thermodiffusion in a multicomponent medium

The liquids appearing in nature and industrial applications are essentially multicomponent. The behavior of multicomponent systems is more complicated in comparison with pure fluids due to a complex interplay between heat and mass transfer processes [18,19]. In mixtures, the diffusive mass transport of a given component is induced not only by its compositional gradient (main or principal diffusion), but also by the compositional gradients of the other components (cross-diffusion) and the temperature gradient.

There are many important processes in nature and technology, where these phenomena play a crucial role. The composition of hydrocarbon reservoirs is significantly affected by diffusion as well as by the Soret effect (due to the presence of geo-thermal gradient) [12,13]. The prediction of mass transfer processes in such systems greatly relies on the knowledge of diffusion and thermodiffusion coefficients, which appear in the equations describing these phenomena.

Kempers [20,21] introduced a new way of calculation for the thermal diffusion factor. His theory was developed for multicomponent mixtures with  $n$  different species. He denotes the mass fraction of component  $i$  by  $C_i$  and obtains the equation giving the mass flux of the constituent  $i$ :

$$J_i = -\rho_0 \sum_{k=1}^n D_{ik} \nabla C_k - \rho_0 C_i (1 - C_i) D_{Ti} \nabla T \quad (8)$$

For a ternary mixture, considering that the two components 1 and 2 are independent, the third one is chosen as a solvent then:

$C_1 + C_2 + C_3 = 1$  and the diffusive fluxes of independent components can be written as:

$$J_1 = -\rho_0 D_{11} \nabla C_1 - \rho_0 D_{12} \nabla C_2 - \rho_0 F_1(C_1, C_2) D_{T1} \nabla T \quad (9)$$

$$J_2 = -\rho_0 D_{21} \nabla C_1 - \rho_0 D_{22} \nabla C_2 - \rho_0 F_2(C_1, C_2) D_{T2} \nabla T \quad (10)$$

Different formulations giving the expression of functions  $F_1(C_1, C_2)$  and  $F_2(C_1, C_2)$  are used. Shevtsova et al. [22] adapted for the ternary mixture the equation obtained by Kempers [17] for multicomponent:

$$F_1(C_1, C_2) = C_1(1 - C_1) \quad \text{and} \quad F_2(C_1, C_2) = C_2(1 - C_2) \quad (11)$$

Ghorayeb and Firoozabadi [23] employed:

$$F_1(C_1, C_2) = C_1(1 - C_1 - C_2) \quad \text{and} \quad F_2(C_1, C_2) = C_2(1 - C_1 - C_2) \quad (12)$$

while Larre et al. [24] used a new form of phenomenological laws:

$$J_1 = -\rho_0 D_{11} \nabla C_1 - \rho_0 D_{12} \nabla C_2 - \rho_0 (C_1 C_2 D_{T12} + C_1 C_3 D_{T13}) \nabla T \quad (13)$$

$$J_2 = -\rho_0 D_{21} \nabla C_1 - \rho_0 D_{22} \nabla C_2 - \rho_0 (C_2 C_1 D_{T21} + C_2 C_3 D_{T23}) \nabla T \quad (14)$$

where  $D_{ij}$  are the mass-based diffusion coefficients and  $D_{Ti}$  are the thermal diffusion coefficients. The transport coefficients are supposed to be constant in a sufficiently small range of temperature and concentration variations and correspond to the mean temperature  $T_0$  and composition  $C_0 = (C_1^0, C_2^0, C_3^0)$ . The diffusive flux of  $J_3$  is defined from the condition that the fluxes of all components must sum up to zero.

The coefficients  $D_{ij}$  and  $D_{Ti}$  can be estimated from theoretical models or measured experimentally. Presently, there is no finished and self-consistent theory for the ternary systems, but there is a hope that such a theory can be formulated using thermodynamic approach and verified by reliable measurements. Nowadays, there are very few experimental studies in ternary liquid mixtures where the thermodiffusion coefficients are determined [25–28]. Until now the values of the Soret coefficient were measured only in three hydrocarbon liquid ternary mixtures. The binary contributions of one among the three hydrocarbon mixture were used as benchmark liquids [29], and it appears that this mixture also plays the role of a benchmark mixture among the ternary ones.

The validity of some empirical additive rule to estimate the thermodiffusion coefficients in a ternary mixture from those in the three corresponding binary mixtures is still under investigation.

#### 4. Measurements of the Soret coefficients

Measurements of Soret coefficients for various binary mixtures have been undertaken since the discovery of the thermodiffusion phenomenon, but this is not an easy task (and measurements are always very accurate) because of the combined effects of the smallness of concentration gradients and the possible undesired convective currents in the liquid bulk.

The oil industry is particularly interested in multicomponent systems, but before starting experiments, even in a ternary system, one has to be sure of the correctness of the measured Soret coefficient in binary systems. The direct determination of  $S_T$  based on Eq. (2) could be delicate, given that it implies the absence of convection, which is not always easy under terrestrial conditions (normal gravity), but convection can be avoided in well-conducted experiments [30,31]. Alternatively, experiments in microgravity have been proposed and performed [32] to alleviate the difficulty that could result from unexpected convective currents.

Several research groups from different universities and research institutes met in Fontainebleau [29] in order to establish a reliable database of Soret, thermodiffusion, and isothermal diffusion coefficients, using different techniques such as thermogravitational cells [30,31] and laser-Doppler velocimetry to analyze the influence of the Soret-induced solutal contribution to the buoyancy force and its consequence on velocity amplitudes [29], or very precise optical techniques such as thermal diffusion forced Rayleigh scattering (TDFRS) [33]. They used binary mixtures of dodecane, isobutylbenzene, and 1,2,3,4-tetrahydronaphthalene (components of interest in the oil industry), the values produced by each group did not differ too much from each other.

Two recent reviews and Ph.D. thesis have summarized the current experimental approach [33–35]. Roughly the experimental methods can be divided into two classes: (i) methods that employ the presence of buoyant convection in the measurements; (ii) methods in which buoyant convection plays a deleterious role and efforts are made to minimize its effect. The major part of the second group is optical methods which rely on the variation of refractive index with composition. The main features of optical methods are non-intrusiveness and sensitivity. A significant advantage of these methods is the absence of mechanically driven parts in contact with the liquid, so measurements do not disturb the experiment. The main disadvantage is that the media under investigation must be transparent. Recent advances in lasers and electronic cameras as well as increasing possibilities of computer-aided data processing have enabled considerable progress in the development of new optical measurement techniques along with a revival of traditional techniques.

In fact, there is no universal working technique acting satisfactorily for every mixture, and each technique has its own limitations.

#### 5. Vibrations and thermodiffusion

Coupling thermodiffusion, natural convection and vibrations is important for applications related to the microgravity. Many works have been devoted to thermo-vibrational convection in fluid and porous media. It is well known that high-frequency accelerations induced by crew activities in microgravity platforms (g-jitter) can produce drastic disturbances during the experiments in space, such as, for instance, in solidification processes during which mushy zones, modeled as porous media, are produced. The g-jitter leads to a non-zero mean flow which may have an important effect on the average heat transfer. High-frequency vibrations can also significantly alter Earth-bound experiments. In the case of weightlessness, only the specific thermo-vibrational mechanism is responsible for instabilities.

All these previous works show that it is important to study the control of convective motions by vibration effects either in a single constituent or a binary fluid or in a porous medium saturated by a single constituent or a binary fluid. Gershuni et al. [36,37], analyzed the stability of mechanical quasi-equilibrium of a binary mixture horizontal layer subjected to a vertical temperature gradient, under a high-frequency vibrational field, when Soret effect is taken into account.

Recently, Shevtsova et al. [38] produced a benchmark of numerical solutions of the vibrational convection problem with the Soret effect in a cubic rigid cell filled with water (90%) and isopropanol and subjected to a temperature gradient between two opposite lateral walls.

Charrier-Mojtabi et al. [39] considered the influence of the direction of vibration on the stability threshold of two dimensional Soret-driven convection in an infinite layer filled with a binary mixture, which can be heated from below or from above. They concluded that the vertical vibration has a stabilizing effect, while the horizontal vibration has a destabilizing effect on the onset of convection.

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