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Review

Perspective of research on diffusion: From microgravity to space exploration

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ABSTRACT

Liquid mixtures are fundamental model systems for the investigation of non-equilibrium processes because they exhibit stable or unstable configurations due to their density stratification, determined by the interplay of their temperature and concentration profiles. The microgravity conditions on platforms like the International Space Station (ISS) provide a unique environment to research multicomponent mixtures under stable conditions, minimizing convective motions. Projects like IVIDIL, SCCO, and GRADFLEX have advanced the understanding of diffusion and thermal diffusion in binary mixtures, triggering the development of specialized research facilities needed for the investigation of multi-component mixtures. The DCMIX project was pivotal in this area because the investigation of thermodiffusion in ternary mixtures demonstrated the importance of microgravity for a precise determination of the transport coefficients. Based on DCMIX's success, the Giant Fluctuations project (also called NEUF-DIX) will investigate diffusion processes in complex fluids at the mesoscopic scale, including polymers, colloids, and proteins. A significant revelation from these space projects is the emergence of the role of phenomena like cross-diffusion and diffusiophoresis in ternary mixtures. In fact, these phenomena are especially pertinent to space exploration, where understanding the stability of liquid mixtures is crucial for the endurance of pharmaceuticals, food ingredients, and construction materials during missions to the Moon and Mars. Control over the stability of these mixtures is essential to prevent phase separation and aggregation in complex fluids and to process these substances in space under defined conditions.

1. Introduction

Liquid mixtures represent an important model system for the investigation of non-equilibrium processes [1]. The non-equilibrium condition is typically represented by the presence of a concentration profile inside the mixture. This concentration profile is associated to a density stratification of the mixture, which in the presence of gravity can be either stable or unstable. In the stable case, the density of the mixture decreases monotonically with height and the system undergoes a purely diffusive process. Conversely, the unstable case corresponds to the configuration where the density profile is top-heavy, which can lead to the onset of a convective instability. The presence of a temperature gradient inside the mixture determines a further control parameter that significantly alters the stability of the mixture. This is due to the fact that the temperature gradient induces a mass flow $j_i = \rho D S_T w_i(1 - w_i)\nabla T$ inside the mixture due to the Soret effect. Here ρ is the density of the mixture, D the diffusion coefficient, w the weight fraction, S_T the Soret coefficient and T the absolute temperature. Under these conditions, the stability of the mixture largely depends on whether the Soret coefficient is positive or negative. The case of a negative Soret coefficient is of particular interest, because when the system is heated from above, a configuration that would be stable in a single component fluid, the Soret effect can determine the formation of a top-heavy density profile, leading to the onset of a solutal convective instability [2].

1.1. The microgravity era of space research

Multicomponent mixtures in the presence and in the absence of thermal gradients are an important model system for the fundamental understanding of how the competition between antagonist effects affects the stability of the system. At the same time, they are largely relevant for practical applications such as the oil recovery and petroleum industry, chemical engineering, the food and beverage industry, the pharmaceutical industry, and material science. During the microgravity era of research in space, the fact that their stability is largely affected by the presence of gravity made them largely investigated

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under reduced gravity conditions on Low Earth Orbit (LEO) platforms such as sounding rockets, unmanned satellites, and platforms like the International Space Station [3]. These platforms allowed to perform experiments under conditions where the transport of mass inside the mixture is entirely determined by diffusion and thermodiffusion processes, in the absence of convective instabilities generated by terrestrial gravity. Within this framework, the IVIDIL project investigated the interaction between diffusion and vibrations in microgravity settings, and revealed that high-frequency periodic forcing leads to time-averaged flows, profoundly impacting diffusive mass transfer [4]. The SCCO project, supported by petrochemical industries such as TOTAL, PETROCHINA, and RIPED, encompassed numerous experiments across various space platforms with the primary objective of examining thermodiffusion in multicomponent mixtures under reservoir thermodynamic conditions relevant to the petroleum industry [5–7]. The GRADFLEX project investigated heat and mass transfer at the mesoscopic scale aboard the FOTON M3 spacecraft, and provided compelling evidence that under microgravity conditions these processes involve significant non-equilibrium fluctuations, limited only by the size of the sample [8,9]. These space experiments allowed to reach a deep understanding of diffusive processes under ideal conditions.

To go beyond these ideal conditions, further complexity can be added to the system by increasing the number of components inside the mixture. On Earth, this additional degree of freedom of the system can determine the onset of convective instabilities even in isothermal systems prepared in the initial condition where the density profile is stable, either through a double diffusion process where one of the solutes has a higher diffusion rate than the other, or through a cross-diffusion process where the concentration gradient of one of the solutes determines a mass flow of the other. For these reasons, experiments on multicomponent mixtures aimed at the determination of their transport coefficients can only be performed under microgravity conditions. This was the case of the DCMIX project backed by ESA and ROSCOSMOS, which aimed to explore thermodiffusion in ternary liquid mixtures through four space missions on the International Space Station from 2012 to 2018. DCMIX enabled a systematic exploration of thermodiffusion in several model ternary mixtures, expanding the Fontainebleu benchmark previously established for binary systems [10] and pioneering the establishment of the first benchmark for ternary mixtures [11].

The DCMIX campaigns allowed to establish the importance of microgravity conditions for the investigation of multicomponent mixtures under non-equilibrium conditions, and laid the grounds for the Giant Fluctuations project (NEUF-DIX) of ESA, which will investigate diffusion processes occurring in complex fluids at the mesoscopic scale. The Giant Fluctuations project envisions 20 different experiments on multicomponent polymer solutions, colloidal suspensions and biological samples under non-equilibrium conditions [12,13].

1.2. The space exploration era

During the last ten years, the priorities of research in space have been largely modified. While microgravity experiments are still deemed important to achieve a fundamental understanding of physical processes affected by gravity, much of the attention has been shifted to the exploration of space.

The new vision for the robotic and manned exploration of space led to the development of the Terranovae 30+ Strategy Roadmap of ESA [14], which aims to continue experiments in Low Earth Orbit, land European astronauts on the Moon by 2030, and send Europeans to Mars by 2040. To identify the new priorities of space research needed for the definition of the Terranovae 30+ program, the European Space Agency collaborated with teams of scientists on the preparation of White Paper summarizing outstanding research topics that could be tackled with Low Earth Orbit platform, or during long term missions on Moon and Mars [15]. The White Paper on Soft Matter and Biophysics covered topics such as diffusion in liquid mixtures, nucleation and structures, soft particles dispersions, granular materials, complex plasmas, active matter, and the physical and biophysical aspects of morphogenesis [16]. Among these, the understanding of diffusion in liquid mixtures in the presence and in the absence of external fields and reactions represents a challenging topic of strategic relevance for space exploration. The investigation of diffusive processes, including thermodiffusion, cross-diffusion and double diffusion, at the mesoscopic scale is becoming increasingly important for the understanding of mass transfer in confined systems [17]. The White Papers served as input to identify outstanding scientific questions. However, addressing all these questions would require a huge amount of resources and is not feasible in practice. For this reason, the European Space Agency collaborated with an interdisciplinary team of scientists to work on the identification of a few Spotlights of general relevance that could represent the backbone of future plans. This led to the identification of two scientific questions of wide relevance in the fields of Soft Matter-Biophysics: (i) How does matter mobilize, move, settle and arrest in space environment and how do these processes affect its transport, processing and storage? (ii) What principles govern the transition from small to large and from short-term to long-term scales in complex systems (such as planetary atmospheres, biological ecosystems, soft matter)?

Within this context, the DCMIX and Giant Fluctuations projects have paved the way for a new era of experiments on liquids of crucial relevance for the exploration of space. Indeed, the understanding of the stability of liquid mixtures under various gravity conditions represents an important strategic factor to guarantee a long shelf life of pharmaceuticals, ingredients for foods, chemicals for fuel and construction materials needed during long term space missions on the Moon and Mars. With this respect, the control of the stability of liquid mixtures would allow to avoid phase separation and aggregation in complex fluids. At the same time, these fluid materials will have to be processed in space when needed, and the ability of controlling mixing in the presence of external fields assume pivotal relevance for long term exploration missions.

In the following we will briefly outline the DCMIX and Giant Fluctuations projects. These two projects have investigated and will investigate in detail thermodiffusion processes in ternary liquid mixtures, both at the microscopic and mesoscopic length scales. An important result of these studies on multicomponent systems is the emergence of a new phenomenology determined by cross-diffusion and diffusiophoresis. In cross-diffusion a gradient in the concentration of one component of the mixture induces a flux of another component, while in diffusiophoresis the concentration gradient of a molecular solute induces a flux of colloidal particles or macromolecules dispersed in a multi component solvent. Both these processes significantly alter the gravitational stability of a multicomponent mixture with respect to the case of a binary one, where rather generally a gravitationally stable mixture remains stable in time, with a few notable exceptions such as the carbonation process. Since multicomponent mixtures under nonequilibrium conditions are ubiquitous in geological and biological systems, we think that the investigation of cross-diffusion and diffusiophoresis represents a new important scientific frontier largely relevant for space exploration, where multicomponent fluids present in organisms, in technological materials, food and in the destinations of the space travels will be subjected to various levels of gravity, and we will dedicate part of this manuscript to the presentation of recent results in these fields.

2. DCMIX as the cornerstone of ternary mixture research

A substantial part of the information regarding thermodiffusion in ternary mixtures available in the literature originates from research carried out within the DCMIX project. This project included conducting measurements of transport coefficients in microgravity aboard the International Space Station (ISS). The European Space Agency (ESA)

played a leading role throughout the project. The Russian space agency was actively involved in the project, while Canada's participation was limited to the initial stage. The experiments on the ISS were carried out between 2012 and 2018 and the DCMIX project was closed by the end of 2021. Four campaigns of the DCMIX experiment were targeting several different ternary mixtures. The systems investigated on the ISS comprised hydrocarbons (DCMIX1) [18], highly nonideal mixtures with demixing zone (DCMIX2) [19], and hydrogen-bonding systems (DCMIX3) [20]. An exploratory campaign (DCMIX4) [21] included three cells of DCMIX2 mixture at compositions closer to the demixing zone and complex fluids; one cell with the solution of C60 fullerene in tetralin–toluene solvent; while another two cells (ternary and binary) fluid consisted of a solution of polystyrene in toluene–hexane solvent. The acquired knowledge indicates the general research direction for investigating ternary mixtures through several pathways. These paths include determining optical contrast factors and the mass diffusion matrix, as well as measuring the Soret coefficients using different techniques. The initial two paths serve as prerequisites for pursuing the third. While the condition number of the optical contrast factors matrix determines the feasible mixture compositions for measurement [22], the diffusion matrix enables obtaining the definitive values of the Soret coefficients. Conducting systematic thermodiffusion experiments in microgravity is constrained by the limited availability of experimental time and the significant costs associated with such efforts. The DCMIX experiments carried out onboard the ISS have stimulated ground-based research, and ternary systems have been studied from a combined experimental, numerical and theoretical perspective.

3. Giant fluctuations: understanding diffusion in complex systems at the mesoscopic scale in microgravity

The ESA's GRADFLEX experiment demonstrated that under microgravity condition, a stationary thermodiffusion process exhibits large non-equilibrium fluctuations, with sizes comparable to the sample itself [9,23,24]. While small concentration gradients can be described using linearized hydrodynamics, these linear models are inadequate for most practical applications and are limited to ideal conditions. Therefore, a key objective of the Giant Fluctuations project of ESA is to explore these fluctuations under non-ideal conditions, such as large gradients, transient processes, and concentrated samples. Additionally, the project aims to understand how these fluctuations influence the interactions between macromolecules, which may be affected by thermal forces resulting from the confinement of non-equilibrium fluctuations [25,26]. The project will investigate non-equilibrium fluctuations in complex fluids, including multicomponent mixtures with a polymer, colloidal suspensions, and protein solutions. To support this research, a dedicated facility has been developed, featuring an array of four shadowgraph optical instruments operating in parallel, each equipped with a thermal gradient cell and a two-color shadowgraph setup [12,13]. Each cell allows to apply to the sample a temperature gradient up to 25 k/mm. The shadowgraphy optical diagnostics illuminates the sample with the collimated violet and red radiation emitted by two super-luminous light sources. Local maxima and minima in the index of refraction determined by temperature and concentration fluctuations give rise to bright and dark regions in the shadowgraph images. The processing of sequences of shadowgraph images allows to attain a fully quantitative statistical characterization of the static and dynamic properties of the fluctuations [27]. Similarly to what has been achieved with two-color interferometry in DCMIX, the utilization of two light sources with well separated wavelengths allows to separate the contributions of the different component of the mixture to the optical signal, due to the different dependence of the components from the wavelength of the radiation. The processing of the large number of images acquired during each experiment will take advantage of innovative parallel algorithms that allow to achieve real time processing [28].

On Earth, the stable density stratification of the mixtures determined by gravity determines a rich phenomenology of the non-equilibrium fluctuations. In fact, long wavelength fluctuations are quenched by the gravity force, which can lead to oscillations and propagating modes that can be also studied using macroscopic model systems [29]. The fact that the amplitude and the relaxation times of the non-equilibrium fluctuations are related to the transport coefficients allows to achieve a reliable determination of the latter by means of a statistical characterization of the fluctuations [30,31]. In the presence of a strong stratification of the mixture, nonlinear behavior can emerge during isothermal diffusion [32] and thermodiffusion [33]. Under these conditions, the distribution of relaxation times of the fluctuations determines a non-exponential relaxation of their time correlation function, requiring the developments of suitable interpretative models where the nonlinear effects are considered. A case of particular interest is represented by binary and ternary mixtures characterized by at least one negative Soret coefficient. In these cases, when heating the sample from above, the unstable solutal contribution to the density gradient competes with the stable thermal contribution, giving rise to nontrivial dynamics. In the case of colloidal suspensions with negative Soret coefficient, this competition between the solutal and thermal mode can give rise to a bistability of the heat transferred [34,35] accompanied by transient localized rotating patterns [36]. In the case of ternary mixtures, two solutal modes with opposite Soret coefficients can also compete and give rise to transient convection [37]. Due to the complex interplay between the thermal and solutal modes, ternary mixtures are characterized by a rich stability diagram and the theoretical description of non-equilibrium becomes challenging. With this respect, a complete theoretical description has been achieved for the nonequilibrium fluctuations in a ternary mixture under the action of a stationary temperature gradient, when the quiescent non-convective state is stable [38].

4. Role of cross-diffusion on emergence of instabilities in ternary mixtures

Most existing research primarily deals with straightforward cases involving regular diffusion. However, cross-diffusion represents a significant phenomenon to understand the dynamic behavior of multicomponent systems. This is due to the fact that it reveals the directional movement of species, and is widely observed in chemical, biological, and physical systems. In perspective, understanding the impact of cross-diffusion on mass transport in mixtures containing multiple components is an important area for future study. In this section, we will briefly discuss recent discoveries regarding cross-diffusion and suggest possible avenues for further research.

In ternary mixtures, three species interact through cross-diffusion, where each species is not only transported by its own concentration gradient, which actually serves as the driving force for transport, but also influenced by the concentration gradients of the other two species. The focus of our research is on the toluene (T)-methanol (M)-cyclohexane (Ch) ternary mixture, studied both on-board the ISS (DCMIX2 mixture) and in ground laboratories, see Fig. 1. The integral studies revealed that depending on composition, the cross-diffusion coefficients $D_{i,j}, i, j = 1,2$ can be as large as the main ones $D_{i,i}, i = 1,2$. This mixture also exhibits the Soret effect and the studies showed that the Soret coefficients of the denser components are negative in a wide range of compositions that result in the hydrodynamic instability in non-isothermal gravity field. In addition, in the region of compositions with large cross-diffusion, the Soret and thermodiffusion coefficients exhibit opposite signs. This is the first mixture where such disparity was found due to on-orbit experiments. Note that in binary mixtures, the signs of the Soret and thermodiffusion coefficients are the same. Our attention lies on the state point **A** with composition T-M-Ch=[(0.62 - 0.31 - 0.07] in mass fractions, which are located on the border of hydrodynamic stability and its surroundings. Such point is characterized by a nearly

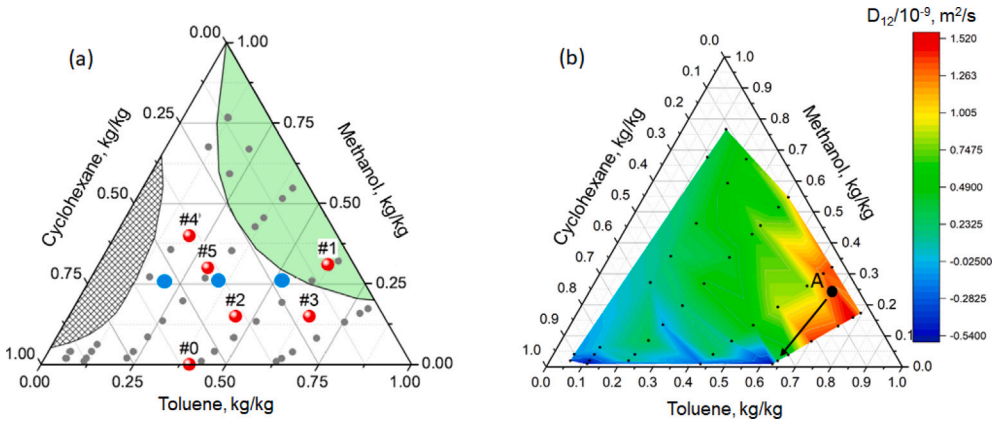

Fig. 1. The toluene-methanol-cyclohexane mixture. (a) Investigated points to detect the Soret effect: during DCMIX2 (red circles) [19,39], during DCMIX4 (blue circles) [21] and in the laboratory (small gray circles) [40]. The shaded area represents the demixing zone. (b) The color map representing the cross-diffusion coefficient D_{12}, important for this study, is derived from the experiments in [41,42]. Present analysis focuses on instabilities at point **A** with composition T-M-Ch=(0.62 - 0.31 - 0.07) kg/kg and its surroundings. The arrow indicates the direction of the major change in D_{12} near point **A**, primarily influenced by variations in methanol content. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

zero net separation ratio Ψ [43], while individual separation ratios ψ_i are typical,

$$\Psi = \psi_1 + \psi_2, \qquad \psi_i = -(\beta_{w_i}/\beta_T) S'_{T,i}, \tag{1}$$

$$\beta_{w_i} = -\frac{1}{\rho}\frac{\partial \rho}{\partial w_i}\bigg|_{T_0,w_0}, \quad \beta_T = -\frac{1}{\rho}\frac{\partial \rho}{\partial T}\bigg|_{T_0,w_0}.$$

Here, β_{w_i} and β_T and are the solutal and thermal expansion coefficients, respectively. $S'_{T,i} = w_i(1-w_i)S_T$ is the modified Soret coefficient. The T-M-Ch system was explored in the vicinity of point **A** under three different conditions. In particular, we discuss the response of the system under thermal gradient conditions, when the Soret effect becomes important, i.e., in a thermogravitational column (TGC) [44,45] and the Soret cell [46]. Under isothermal conditions, the diffusion of two layers of a mixture of slightly different composition may result in fascinating pattern formation, influenced by both diffusion and cross-diffusion processes [47]. In the gravitational field, diffusion can cause convective patterns in an initially stable system. In this case, diffusion induces interfacial destabilization either when the component of the lower layer diffuses faster than the upper one, or vice versa. The most well-known example describing a double diffusive (DD) instability is when warmer salt water is over cold fresh water and salt fingers can occur. The presence of cross-diffusion complicates the phenomenon. In each of these three cases, the emerging instability exhibits intriguingly distinct behavior.

4.1. Thermogravitational column (TGC)

The experiments with ternary mixtures were conducted using the TGC, revealing oscillatory behavior that remained unexplained for several months. The absence of supporting numerical studies was primarily due to the lack of data on diffusion and especially thermodiffusion coefficients, since the latter can only be measured in a stable system. This problem has been resolved thanks to the recent DCMIX experiments conducted in space, with particular emphasis on the DCMIX2 experiment relevant to this study [19].

The TGC is a side-heated narrow vertical slot, the sketch of it is shown in Fig. 2. The horizontal separation of components due to the Soret effect combined with the vertical convective current leads to the appearance of vertical concentration gradients. These gradients reach constant values at the steady state. The establishment of a stable vertical distribution of heavier components allows the measurement of thermodiffusion coefficients [48]. However, the hydrodynamic stability of the system can be violated depending on the characteristics of the mixture, in particular on the separation ratio [49]. Previous theoretical

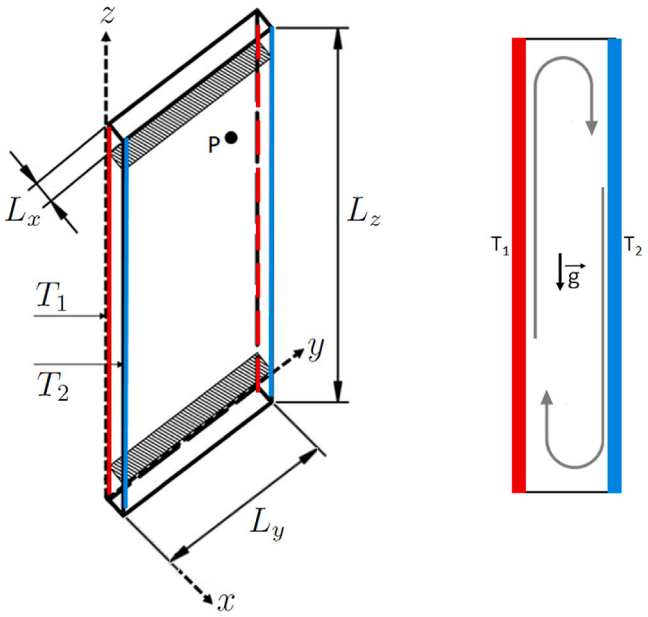

Fig. 2. The geometry of thermogravitational column and sketch of its working principle. The liquid is placed in a narrow vertical column, the two opposite walls of which are maintained at different temperatures. The presence of a horizontal temperature gradient causes the Soret effect, leading in the separation of components along the horizontal axis and transported in vertical direction by convection.

studies [50–52] have shown that instabilities can occur in ternary mixtures regardless of whether the net separation ratio (Ψ) is positive or negative. However, the precise nature of instability depends on the combination of the ψ_i signs of the individual components in the mixture.

The ground experiments [44], followed by numerical simulations using the coefficients measured on-orbit [44,45], have revealed the existence of longwave instability in the M-T-Ch mixture. It was elucidated that a large cross-diffusion is responsible for its occurrence. Further analysis has unveiled the emergence of a secondary instability in the form of a swinging pattern, alongside the dominant large-scale standing wave.

Fig. 3 illustrates these results and provides evidence of the instability occurring in the transverse direction in the form of a standing wave. The system requires some time to attain sustained periodic

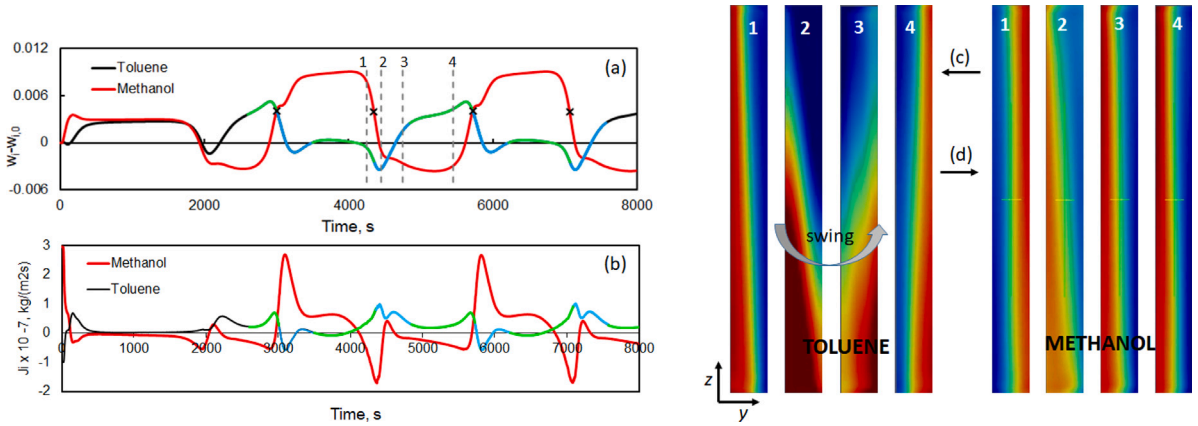

Fig. 3. (Numerical results [44].) Oscillatory behavior of (a) mass fractions $w_i^* = (w_i - w_{i0})$ and (b) vertical net mass fluxes J_{iz} of toluene and methanol at point P (shown in Fig. 2) at $D_{12} = 1.34 \cdot 10^{-9}$ m^2/s. The green color on the curves for w_1, J_1 illustrates the diffusion phase of a standing wave. The blue color on curves indicates the regions with the occurrence of swinging instability. Panels (c) and (d) present the evolution of mass fractions w_i^* over the half of a period in transversal direction, consisting of two phases: fast and slow. To highlight the disparity in the duration different phases, the snapshots at t = 4250, 4450, 4650 and 5450 s are not equidistant. The timing of snapshots is shown by vertical gray lines in panel (a). The first and fourth snapshots constitute half of the period. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

behavior, during which the instability gradually sets in. The waveforms of curves for toluene (w_1) and methanol (w_2) are noticeably different, with methanol (red curve) clearly displaying two phases of oscillations. These phases consist of long intervals where the mass fraction (w_2^*) remains practically unchanged (e.g. at 3400 s < t <4000 s), followed by shorter intervals characterized by rapid changes. The observed rapid changes are associated with swinging instability, which occurs within the context of a standing wave. The latter takes a longer time to establish. To highlight different instability modes, the regions of occurrence of swinging instability are shown in blue on the w_1, J_1 curves, and the diffusion phase of a standing wave is illustrated in green.

The feature of a standing wave is the change of rich/poor areas of analyzed quantity every half of a period. The existence of a standing wave in the considered system is confirmed by the presence of red and blue colors on opposite sides in the first and fourth snapshots (panels (c) and (d)), which constitute half of the period. The difference in the patterns between the toluene and methanol shown in 2nd and 3rd snapshots indicates that the toluene drives the swinging instability. Furthermore, a strong contribution of cross-diffusion effect (D_{12}) causes the toluene flux to be opposite to the methanol flux (see panel b), and this leads to the development of instability. Thus, the large cross-diffusion D_{12} leading to a mismatch in signs between Soret and thermodiffuion coefficients, is responsible for a swinging motion of w_1 pattern, and, in addition, this is the motor of the entire oscillatory instability.

Cross-diffusion also plays an important role in determining the oscillation period. For the state point **A** with $D_{12} = 1.3 \cdot 10^{-9}$ m^2/s (see Fig. 1)(b), the numerical simulations [44,45] yielded a period of Π = 2740 s, while experiments [44] exhibited $\Pi \approx 4000$ s. It was noticed that the oscillation period increases significantly with decreasing D_{12}. Correspondingly, the discrepancy between experimental and numerical values was attributed to the inherent challenges in accurately measuring cross-diffusion coefficients. Subsequent studies have shown that this difference can be explained by the dependence of cross-diffusion values on composition of a mixture.

4.1.1. Concentration-dependent cross-diffusion

To further highlight the role of cross-diffusion, we present new results obtained by conducting simulations incorporating composition-dependent cross-diffusion. We utilized the numerical code developed in [44,45]. In order to isolate the effect of cross-diffusion, the remaining parameters of the system are considered constant. The simulations were carried out for different amplitudes of D_{12} variation while maintaining the mean value constant, and for several mean values

$$D_{12} = \alpha\, w_2 + \gamma \tag{2}$$

The coefficients α and γ are selected in such a way (see Table 1) that this equation reflects the dependence observed in the real experimental data shown in Fig. 1(b). The cross-diffusion coefficient essentially changes in the direction indicated by the arrow, and this change is primarily affected by the methanol content. The set of parameters used in the simulations is listed in Table 1. The w_2 mass fraction changes around its original value of w_2 = 0.31 kg/kg depending on the considered amplitude of D_{12} variation. This range of w_2 values is outlined on the Gibbs triangle in Fig. 1(b).

Fig. 4 illustrates the impact of variable cross-diffusion on the emergence of instability within the system. The dotted vertical lines indicate the analyzed amplitude of variation of D_{12} at given mean value of D_{12}. The blue inclined line marks the region where oscillatory instability is observed at a constant D_{12}. From these new results, two conclusions can be drawn:

(1) Variable cross-diffusion extends the range of oscillatory regime. Previous findings [44] had shown that under constant cross-diffusion coefficients, oscillatory instability occurs only within a limited range of the D_{12} values, i.e. $1.15 \cdot 10^{-9}$ m^2/s < D_{12} < $2.1 \cdot 10^{-9}$ m^2/s. Beyond this range, the system transitions to a stationary pattern of either Turing-like or monotonic instability. The intriguing behavior is observed considering new cases 1-4 in Fig. 4. While under constant D_{12} or its small variations, this region corresponds to Turing like instability, when the amplitude of D_{12} is large (as depicted by the green line in case 2) the system is capable of exciting oscillations.

Table 1
List of cases to study the influence of variable diffusion: the mean value $\bar{D}_{12}$, the arbitrary coefficients α and γ in Eq. (2), the amplitude of variation ΔD_{12} and period of oscillations Π, when available.

#	$\bar{D}_{12}/10^{-9}$ m^2/s	$\alpha/10^{-9}$ m^2/s	$\gamma/10^{-9}$ m^2/s	$\Delta D_{12}/10^{-9}$ m^2/s	Π s
1	1.0	16	3.96	0.8–1.2	No Osc
2	1.05	50	14.45	0.425–1.675	4500
2a	1.05	−50	1.655	1.675–0.425	4500
3	1.05	16	3.91	0.85–1.25	No Osc
4	1.08	18	4.505	0.85–1.3	No Osc
5	1.10	20	5.1	0.85–1.35	6000
6	1.30	24	6.14	1.0–1.6	2800
7	1.30	48	13.58	0.7–1.9	3000
8	1.9	32	8.02	1.5–2.3	1200

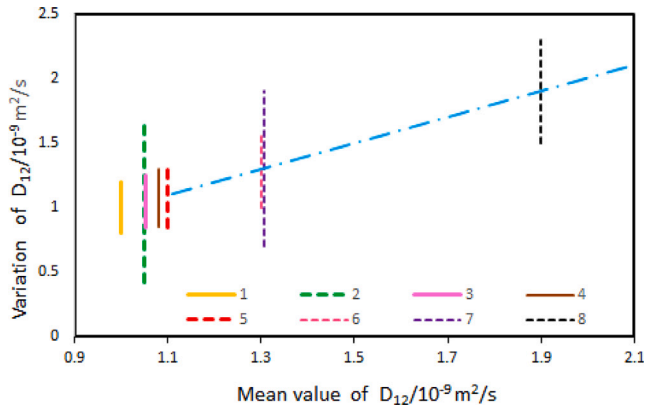

Fig. 4. Impact of variable cross-diffusion on emerging instability. The vertical lines show the amplitude of D_{12} variation at given mean value of $\bar{D}_{12}$, among which the dashed lines corresponds to oscillatory regime and solid line to the Turing like instability. The blue dash-dotted line marks the region where oscillatory instability is observed at a constant cross-diffusion coefficient. The exact values of parameters are listed in Table 1.

(2) The period of oscillations depends on the amplitude of D_{12} variations. For example, for constant $D_{12} = 1.3 \cdot 10^{-9}$ m^2/s, the computed oscillation period is $\Pi = 2740$ s and for the same mean value but for small amplitude variation (case 6) $\Pi = 2800$ s and for larger amplitude variation (case 7) $\Pi = 3000$ s approaching to the experimental value $T \approx 4000$ s [44]. This dynamics is more evident at smaller D_{12} where the oscillation period showed strong increase on its variation even at constant value. For example, for $\bar{D}_{12} = 1.1 \cdot 10^{-9}$ m^2/s (case 5), the period is $\Pi = 6000$ while for extreme value of constant where oscillation existed ($\bar{D}_{12} = 1.15 \cdot 10^{-9}$ m^2/s), the period was $\Pi \sim 4000$ s.

4.2. Soret cell

Experiments and numerical simulations for the same T-M-Ch system were carried out in the Soret cell geometry [46]. Unlike observations using TGC, the system behavior in the Soret cell shows the prevailing effect of the small net separation ratio over cross-diffusion. The system dynamics depends on initial conditions or external perturbations, revealing metastable behavior. In the case of weak perturbations, the mixture remains motionless, facilitating the measurement of transport coefficients, while strong perturbations lead to the development of convective flow [46]. Fig. 5 illustrates these observations. In panel (a), the separation of component Δw_1 due to the Soret effect is depicted. Under weak perturbations (lower branch), the Soret separation reaches a certain asymptotic value, enabling the determination of the Soret coefficient. Conversely, under strong perturbations (upper branch), stationary convection ensues, and the component separation diminishes to the level of background noise. The emergence of convective flow can be attributed to local density variations along the gravitational field, facilitated by the initial perturbation. Panels (b) and (c) illustrate density variations across the cell for the first two components, elucidating this behavior.

In the absence of disturbances, both profiles $\Delta\rho_{w1}$ and $\Delta\rho_{w2}$ are linear, but have opposite slopes, as shown in panel (b). They effectively cancel each other out along the entire height of the cell, resulting in a sum close to zero. Therefore, the hydrodynamic stability of the system is primarily determined by the thermal field. In the perturbed solution, panel (c), the stationary convection flow sets in the system, and the density distribution becomes non-monotonic with height. The overall density variation (red curve) is much smaller than individual contributions, but is also affected by flow. The resulting convection excludes the measurement of Soret coefficients [46].

4.3. Isothermal diffusion

In this section, we briefly discuss the behavior of the same ternary system with large cross-diffusion under isothermal conditions with the aim of providing a perspective about new possible research routes. The diffusion process was studied between two liquids, forming a two-layered system with a well-defined interface between them [47]. The initial difference in mass fraction between the layers is denoted as $\Delta w_i = (w_{i,\text{bottom}} - w_{i,\text{top}})$. Starting from a less dense solution above a denser one, two types of diffusion instabilities can be identified. In the case of double-diffusive convection (DD), a distinctive finger-like pattern emerges, significantly disrupting the diffusive interface. Conversely, in the case of diffusion-layer convection (DLC), the initial contact line maintains its sharp and narrow profile for a considerably longer duration compared to what would be expected from simple diffusion.

The presence of cross-diffusion can drive an alternative path to buoyancy-driven convection in an initially stable system. An example of such a scenario in the T-M-Ch ternary system is shown in Fig. 6. The lower part of the graphs corresponds to an instability pattern according to the Rayleigh–Taylor mechanism, which is delimited from the rest of the space by the neutral stability curve corresponding to the equation:

$$\Delta\rho = \Delta\rho_1 + \Delta\rho_2 = \Delta w_1 \beta_{w1} + \Delta w_2 \beta_{w2} = 0. \quad (3)$$

This curve can be determined using solutal expansions alone, without the need to solve diffusion equations [47]. Panel (a) illustrates instability patterns in the absence of cross-diffusion, showcasing the formation

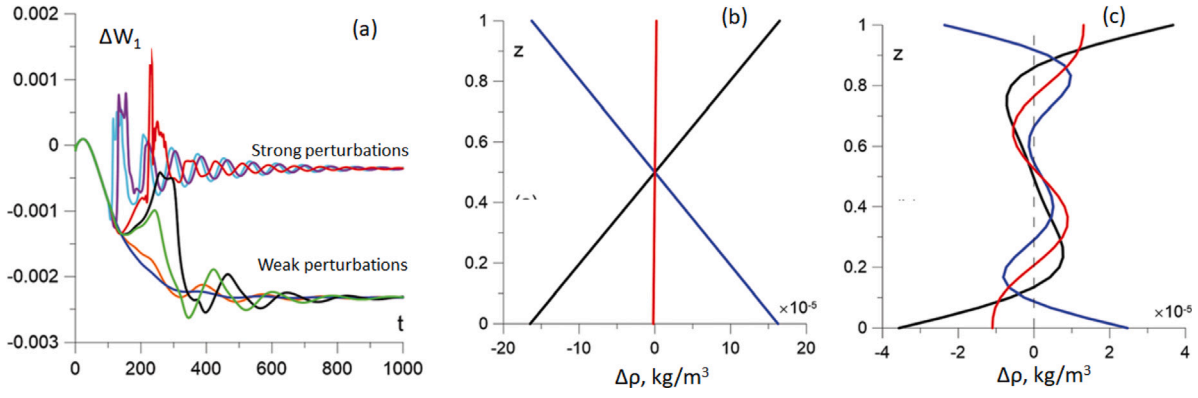

Fig. 5. (a) Time evolution of heavier component (methanol) separation between hot and cold wall due to the Soret effect in the case of weak and strong perturbations of the system introduced in the form of a given vorticity value at one point at t = 0; (b,c) Changes in the density across the cell at = 1000 s caused by the Soret effect in the first component $\Delta\rho_{w1}$ (blue), in the second component $\Delta\rho_{w2}$ (black curve) and their sum $\Delta\rho = \Delta\rho_{w1} + \Delta\rho_{w2}$ (red curve). (b) no initial perturbations;(c) with initial perturbation. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

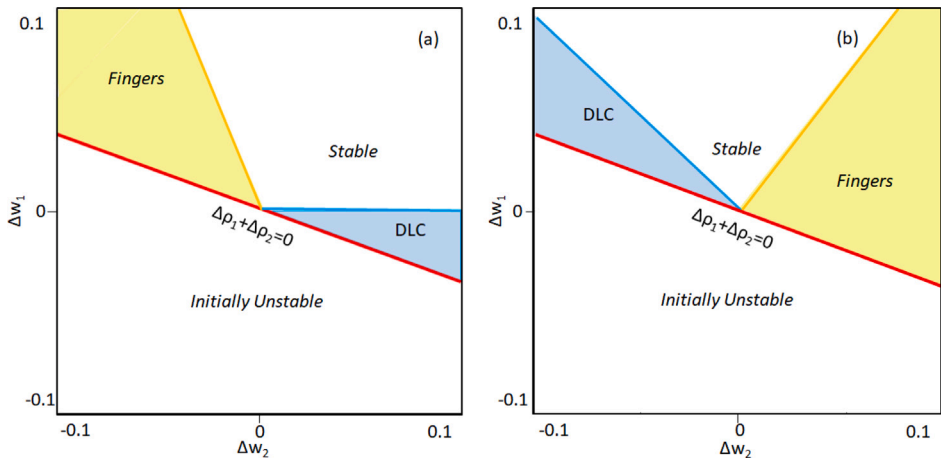

Fig. 6. The impact of cross-diffusion on the instability patterns occurring in the T-M-Ch system at point **A**. The map of states (either fingers or diffusion layer convection) in a ternary system (a) without cross-diffusion; (b) with cross-diffusion. The presence of cross-diffusion switches the location of instabilities between left and right sides.

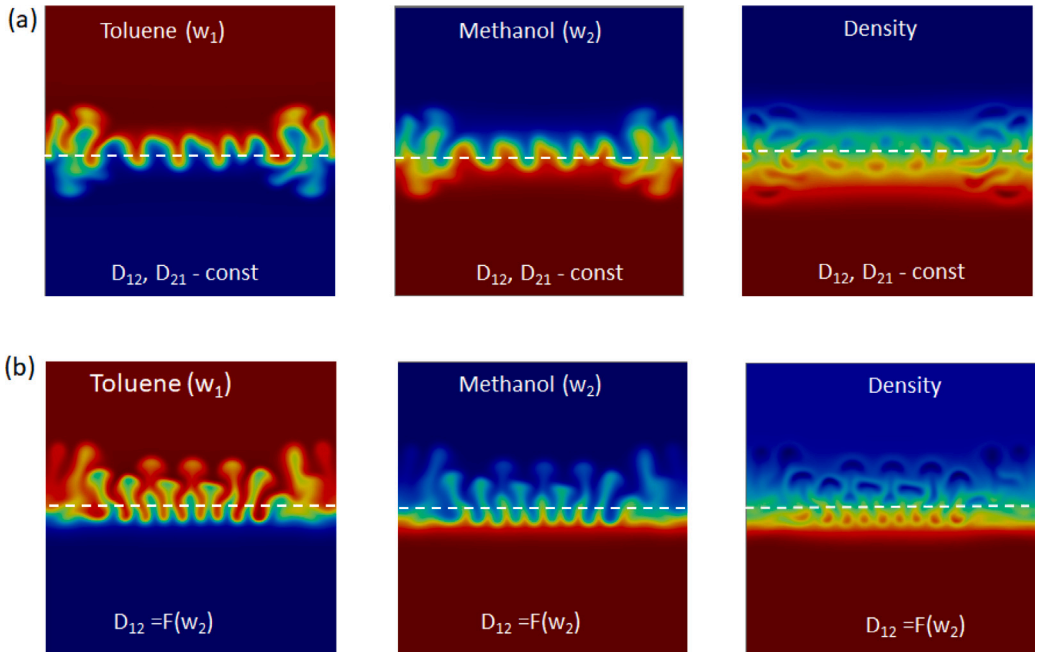

Fig. 7. The influence of constant versus variable cross-diffusion on the emerging diffusion patterns shown through the evolution of the toluene and methanol mass fractions and densities in the T-M-X system a) Cross-diffusion coefficients are constant; (b) D_{12} varies according to Eq. (2). The pictures are taken at t = 600 s; the mixture composition corresponds to the end point of the arrow in Fig. 1(b) (between green and blue regions); the white lines indicate the initial position of the interface. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

of fingers in a large region on the left part of the composition space and diffusion-layer convection (DLC) in a smaller region on the right. In panel (b), a similar map is presented with cross-diffusion taken into account. The instability patterns switch locations: the region of DLC is on the right, while the increased region of fingers (DD) is on the left.

4.3.1. Concentration-dependent cross-diffusion

In this section we present new results aimed at suggesting potential research directions and highlighting analogies in dynamics of systems with cross-diffusion and chemical reactions. We use the same numerical code as in Ref. [47]. The inclusion of composition-dependent cross-diffusion further enriches the variety of observed patterns. In this context, the variable cross-diffusion was incorporated into simulations using the same approach as in the case with temperature gradients case, i.e. according to Eq.(2).

Fig. 7 illustrates the contrasting instabilities resulting from constant and variable cross-diffusion scenarios. Panel (a) gives insight into the dynamic diffusion behavior in a system characterized by constant cross-diffusion coefficients. The mass fraction patterns show symmetric fingers (DD) crossing the initial contact line, creating a wavy structure in the spatial domain. The central fingers exhibit comparable amplitudes, which gradually increase towards the sidewalls. The observed convection appears to be quite powerful, and the effects of convective mass transfer are clearly visible in the density picture.

The diffusion patterns in the system with variable cross-diffusion display distinct characteristics. First, the mass fraction patterns appear as two layers of fingers, each with differing amplitudes and featuring mushroom-shaped heads. Second, these fingers propagate predominantly in one direction — upwards. Despite this upward growth, the lower edge of the fingers intersects the initial contact line and descends.

A more detail analysis shows that this pattern emerges due to the convergence of two depleted zones of low density around the interface. Such a phenomenon has been observed in systems with chemical reactions and downward displacement was dubbed as a shockwave [53].

This 'shockwave' comprises two density levels separated by a thin transition region and progresses towards a denser lower layer.

The observation of the downward movement of the lower edge of the fingers in the ternary system with variable cross-diffusion is particularly intriguing. It underscores the parallels between such ternary systems and reaction–diffusion systems concerning the emergence of instabilities. This phenomenon mirrors aspects of reaction–diffusion dynamics where similar patterns arise from the interplay of diffusive and reactive processes. The resemblance suggests a common underlying mechanism governing the evolution of complex spatial structures in both types of systems.

5. Role of diffusiophoresis on emergence of instabilities in colloidal suspensions

In the presence of a concentration gradient of the solvent determined by a molecular solute, colloidal particles move in the direction of the gradient, a phenomenon known as diffusiophoretic flow, which represents a cross-diffusion effects acting on macromolecules and colloidal particles. Diffusiophoresis has several similarities with thermophoresis and it has been proposed that both these two phoretic mechanisms are driven by an interfacial gradient at the surface of the colloid [54]. The diffusiophoretic drift is not easily explained by classical mechanics and hydrodynamics because, theoretically, the particles should remain stationary due to the absence of a global force and due to the no-slip condition imposed by classical fluid dynamics at the solid–liquid interface of each particle [55].

Microscopic theoretical models of diffusiophoresis indicate that the finite thickness of the colloid-solvent interface results in a diffusio-osmotic process within this layer. This process causes a solvent flow at the particle surface, leading to particle migration in the opposite direction [55–57]. Interestingly, the microscopic models are compatible with a coarse-grained macroscopic description of the process when the size ratio between the solvent and colloidal particles is small [58]. While phoretic processes like electrophoresis and thermophoresis have given rise to well-established technological tools for studying and characterizing macromolecules, diffusiophoresis is a relatively new method, recently gaining attention for technological applications such as microfluidic colloid separation [59] and three-dimensional particle manipulation [60]. Furthermore, the ability to modify the surface of Janus nanoparticles to promote a local solvent reaction has shown potential for developing self-propelled swimmers driven by a local solvent concentration gradient [61–63].

To date, most research on diffusiophoretic processes has focused on understanding the particle motion by modeling the hydrodynamic processes in the thin Debye layer surrounding them. However, there has been limited exploration of emergent phenomena resulting from diffusiophoresis at the macroscopic scale. Understanding the gravitational stability of colloidal suspensions is becoming increasingly important due to their numerous industrial applications and the strategic need to handle and process complex fluids under various gravity conditions for long-term human space missions to destinations like Mars and the Moon [17]. While convective instabilities induced by other phoretic processes, such as thermophoresis, have been extensively studied [64–69], the investigation of convective instabilities induced by diffusiophoresis has been limited. Within this context, recently diffusiophoresis has been shown to play an important role in reaction–diffusion processes, because it provides a mechanism for the formation and sharpening of concentration gradients, which are fundamental to the generation of patterns in biological systems [70]. In the context of Turing patterns, which are reaction–diffusion systems proposed by Alan Turing to explain pattern formation in biological systems, diffusive transport mechanisms have traditionally been relied upon to generate these concentration gradients. However, the diffusive mechanism alone often suffers from poor robustness due to various complicating factors [71]. Diffusiophoresis offers a more robust and effective alternative due to the fact that the propulsion of colloids by a chemical gradient promotes the sharpening of color observed in natural patterns and introduces an independent control parameter for this sharpness, thereby improving the robustness of the models used to simulate Turing patterns. Furthermore, diffusiophoresis is observed to operate effectively under conditions of high Péclet numbers, where convective transport dominates over diffusive transport. For these reasons, diffusiophoresis represents a fundamental process to consider when studying biological pattern formation and other phenomena governed by reaction–diffusion mechanisms.

6. Conclusions and further perspectives

As humanity ventures further into space with the goal of establishing long-term missions on planets like the Moon and Mars, understanding the behavior of fluids and particles in low-gravity and hypergravity environments becomes crucial. In this context, cross-diffusion and diffusiophoresis emerge as phenomena with promising implications for both scientific exploration and practical applications.

Similarly to what happened for separation methods based on thermodiffusion, cross-diffusion holds the potential to improve molecular separation techniques under isothermal conditions. By enhancing the efficiency of separating multi-component mixtures, cross diffusion can significantly improve the capabilities of filtration systems. This advancement could enable the precise separation of various molecular species, with potential applications to water purification and the separation of complex chemical mixtures on Earth and in space. Moreover, cross-diffusion can play a pivotal role in innovating material processing techniques. By enabling more precise control over the distribution and segregation of molecular species, processing methods based on cross diffusion could lead to breakthroughs in industries such as pharmaceuticals, chemicals, and environmental engineering, facilitating the development of new materials and processes tailored for specific applications.

Diffusiophoresis offers unique advantages for the development of technologies suitable for space exploration. It can potentially be used for microfluidic separation of colloids and three-dimensional manipulation of particles, critical for resource extraction and recycling on space missions. In particular, the insights gained from investigating convective instabilities induced by diffusiophoresis could offer valuable information for designing effective fluid management systems in space. Furthermore, the ability to design Janus nanoparticles with specific surface properties opens the door to developing self-propelled swimmers, which can navigate and interact with their environment driven solely by local solvent concentration gradients.

In conclusion, cross-diffusion and diffusiophoresis hold a practical relevance for the success of long-term space missions, and can contribute to the development of innovative technologies, materials, and strategies that are essential for sustaining life and ensuring the success of human exploration beyond Earth. Thus, investing in research and exploration of these phenomena is not just a scientific endeavor but a strategic necessity for the future of space exploration.

CRediT authorship contribution statement

A. Vailati: Writing – review & editing, Writing – original draft, Methodology, Investigation, Funding acquisition, Conceptualization. **B. Šeta:** Writing – review & editing, Visualization, Software, Investigation, Conceptualization. **M.M. Bou-Ali:** Writing – review & editing, Methodology, Investigation, Funding acquisition. **V. Shevtsova:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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References

[1] J.K. Platten, J.C. Legros, Convection in Liquids, Springer, Berlin, Germany, 2011.
[2] R. Cerbino, A. Vailati, M. Giglio, Fast-onset Soret-driven convection in a colloidal suspension heated from above, Phil. Mag. 83 (17–18) (2003) 2023–2031.
[3] M. Braibanti, et al., European Space Agency experiments on thermodiffusion of fluid mixtures in space, Eur. Phys. J. 42 (86) (2019).
[4] V. Shevtsova, Y.A. Gaponenko, V. Sechenyh, D.E. Melnikov, T. Lyubimova, A. Mialdun, Experimental and theoretical study of vibration-induced thermal convection in low gravity, J. Fluid Mech. 767 (290) (2015).
[5] J.C. Legros, S. Van Vaerenbergh, Y. Decroly, F. Montel, Expériences en microgravité étudiant l'effet Soret: SCM, SCCO et MBIS, Entropie 198/199 (1994) 1.
[6] V. Vaerenbergh, S. Srinivasan, M.Z. Saghir, Thermodiffusion in multicomponent hydrocarbon mixtures: Experimental investigations and computational analysis, J. Chem. Phys. 131 (2009) 114505.
[7] G. Galliero, H. Bataller, J.-P. Bazile, J. Diaz, F. Croccolo, H. Hoang, R. Vermorel, P.-A. Artola, B. Rousseau, V. Vesovic, M.M. Bou-Ali, J.M. Ortiz de Zárate, S. Xu, K. Zhang, F. Montel, A. Verga, O. Minster, Thermodiffusion in multicomponent n-alkane mixtures, npj Microgravity 3 (1) (2017).
[8] C.J. Takacs, et al., Thermal fluctuations in a layer of liquid CS2 subjected to temperature gradients with and without the influence of gravity, Phys. Rev. Lett. 106 (2011) 244502.
[9] A. Vailati, et al., Fractal fronts of diffusion in microgravity, Nature Commun. 2 (290) (2011).
[10] J.K. Platten, et al., Benchmark values for the Soret, thermal diffusion and diffusion coefficients of three binary organic liquid mixtures, Philos. Mag. 83 (1965).
[11] M.M. Bou-Ali, A. Ahadi, D.A.d. Mezquia, Q. Galand, M. Gebhardt, O. Khlybov, W. Köhler, M. Larrañaga, J.C. Legros, T. Lyubimova, A. Mialdun, I. Ryzhkov, M.Z. Saghir, V. Shevtsova, S.V. Vaerenbergh, Benchmark values for the Soret, thermodiffusion and molecular diffusion coefficients of the ternary mixture tetralin+isobutylbenzene+n-dodecane with 0.8-0.1-0.1 mass fraction, Eur. Phys. J. E 38 (4) (2015) 30.
[12] P. Baaske, H. Bataller, M. Braibanti, M. Carpineti, R. Cerbino, F. Croccolo, A. Donev, W. Köhler, J.M. Ortiz de Zárate, A. Vailati, The NEUF-DIX space project - non-equilibrium fluctuations during diffusion in complex liquids, Eur. Phys. J. E 39 (12) (2016).
[13] A. Vailati, P. Baaske, H. Bataller, S. Bolis, M. Braibanti, M. Carpineti, R. Cerbino, F. Croccolo, J.-L. Dewandel, A. Donev, L. García-Fernández, F. Giavazzi, R. Haslinger, S. Hens, M. Knauer, W. Köhler, E. Kufner, J.M. Ortiz de Zárate, J. Peeters, C.J. Schwarz, I. Silkina, S. Xu, D. Zapf, Giant fluctuations induced by thermal diffusion in complex liquids, Microgravity Sci. Technol. 32 (5) (2020) 873–887.
[14] European Space Agency, Terranovae 2030+ strategy roadmap, 2022, https://esamultimedia.esa.int/docs/HRE/Terrae_Novae_2030+strategy_roadmap.pdf, (Accessed 22-03-2024).
[15] European Space Agency, The SciSpacE white papers — esa.int, 2021, https://www.esa.int/Science_Exploration/Human_and_Robotic_Exploration/Research/The_SciSpacE_White_Papers, (Accessed 22-03-2024).
[16] European Space Agency, Soft matter and biophysics roadmap, 2021, https://esamultimedia.esa.int/docs/HRE/04_Physical_Sciences_Soft-Matter-Biophysics.pdf, (Accessed 22-03-2024).
[17] A. Vailati, H. Bataller, M.M. Bou-Ali, M. Carpineti, R. Cerbino, F. Croccolo, S.U. Egelhaaf, F. Giavazzi, C. Giraudet, G. Guevara-Carrion, D. Horváth, W. Köhler, A. Mialdun, J. Porter, K. Schwarzenberger, V. Shevtsova, A. De Wit, Diffusion in liquid mixtures, npj Microgravity 9 (1) (2023).
[18] Q. Galand, S. Van Vaerenbergh, W. Köhler, O. Khlybov, T. Lyubimova, A. Mialdun, I. Ryzhkov, V. Shevtsova, T. Triller, Results of the DCMIX1 experiment on measurement of Soret coefficients in ternary mixtures of hydrocarbons under microgravity conditions on the ISS, J. Chem. Phys. 151 (13) (2019) 134502.
[19] A. Mialdun, I. Ryzhkov, O. Khlybov, T. Lyubimova, V. Shevtsova, Measurement of Soret coefficients in a ternary mixture of toluene-methanol-cyclohexane in convection-free environment, J. Chem. Phys. 148 (4) (2018) 044506.
[20] T. Triller, H. Bataller, M. Bou-Ali, M. Braibanti, F. Croccolo, J. Ezquerro, Q. Galand, J. Gavaldá, E. Lapeira, A. Laverón-Simavilla, T. Lyubimova, A. Mialdun, J. Zárate, J. Rodríguez, X. Ruiz, I. Ryzhkov, V. Shevtsova, S. Vaerenbergh, W. Köhler, Thermodiffusion in ternary mixtures of water/ethanol/triethylene glycol: First report on the DCMIX3-experiments performed on the international space station, Microgravity Sci. Technol. 30 (2018) 295–308.
[21] A. Mialdun, H. Bataller, M.M. Bou-Ali, M. Braibanti, F. Croccolo, A. Errarte, J.M. Ezquerro, J.J. Fernandez, Y. Gaponenko, L. Garcí-Ferández, J. Rodríguez, V. Shevtsova, Preliminary analysis of diffusion coefficient measurements in ternary mixtures 4 (DCMIX4) experiment on board the international space station, Eur. Phys. J. E 42 (2019) 87.
[22] V. Shevtsova, V. Sechenyh, A. Nepomnyashchy, J. Legros, Analysis of the application of optical two-wavelength techniques to measurement of the Soret coefficients in ternary mixtures, Phil. Mag. 91 (26) (2011) 3498–3518.
[23] R. Cerbino, Y. Sun, A. Donev, A. Vailati, Dynamic scaling for the growth of non-equilibrium fluctuations during thermophoretic diffusion in microgravity, Sci. Rep. 5 (2015) 14486.
[24] F. Croccolo, C. Giraudet, H. Bataller, R. Cerbino, A. Vailati, Shadowgraph analysis of non-equilibrium fluctuations for measuring transport properties in microgravity in the GRADFLEX experiment, Microgravity Sci. Technol. 28 (2016) 467.
[25] T.R. Kirkpatrick, J.M. Ortiz de Zárate, J.V. Sengers, Giant Casimir effect in fluids in nonequilibrium steady states, Phys. Rev. Lett. 110 (2013) 235902.
[26] T.R. Kirkpatrick, J.M. Ortiz de Zárate, J.V. Sengers, Nonequilibrium casimir-like forces in liquid mixtures, Phys. Rev. Lett. 115 (2015) 035901.
[27] F. Croccolo, D. Brogioli, A. Vailati, M. Giglio, D.S. Cannell, Nondiffusive decay of gradient-driven fluctuations in a free-diffusion process, Phys. Rev. E 76 (4) (2007) http://dx.doi.org/10.1103/PhysRevE.76.041112.
[28] M. Norouzisadeh, M. Chraga, G. Cerchiari, F. Croccolo, The modern structurator: increased performance for calculating the structure function, Eur. Phys. J. E 44 (12) (2021).
[29] M. Carpineti, F. Croccolo, A. Vailati, Levitation, oscillations, and wave propagation in a stratified fluid, Eur. J. Phys. 42 (5) (2021) 055011.
[30] D. Zapf, W. Köhler, Thermal and solutal non-equilibrium fluctuations in a polymer solution, J. Chem. Phys. 153 (22) (2020).
[31] A.T. Ndjaka, L. García-Fernández, D.E.B. Bouyou, A. Lassin, M. Azaroual, F. Croccolo, H. Bataller, Mass diffusion and Soret coefficient measurements of triethylene glycol/water binary mixtures by dynamic shadowgraphy, Eur. Phys. J. E 45 (3) (2022).
[32] S. Castellini, M. Carpineti, C. Giraudet, F. Croccolo, A. Vailati, Dynamics of non-equilibrium concentration fluctuations during free-diffusion in highly stratified solutions of glycerol and water, J. Chem. Phys. 158 (24) (2023).
[33] D. Zapf, J. Kantelhardt, W. Köhler, Nonlinearities in shadowgraphy experiments on non-equilibrium fluctuations in polymer solutions, Eur. Phys. J. E 45 (4) (2022).
[34] G. Donzelli, R. Cerbino, A. Vailati, Bistable heat transfer in a nanofluid, Phys. Rev. Lett. 102 (10) (2009).
[35] M. Bernardin, F. Comitani, A. Vailati, Tunable heat transfer with smart nanofluids, Phys. Rev. E 85 (6) (2012).
[36] M. Carpineti, S. Castellini, A. Pogliani, A. Vailati, Transient localized rotating structures in a suspension of highly thermophilic nanoparticles, Front. Phys. 10 (2022).
[37] L. García-Fernández, H. Bataller, P. Fruton, C. Giraudet, A. Vailati, F. Croccolo, Stabilized convection in a ternary mixture with two Soret coefficients of opposite sign, Eur. Phys. J. E 45 (6) (2022).
[38] J.M. Ortiz de Zárate, L. García-Fernández, H. Bataller, F. Croccolo, Non-equilibrium fluctuations in a ternary mixture subjected to a temperature gradient, J. Stat. Phys. 181 (1) (2020) 1–18.
[39] A. Mialdun, V. Shevtsova, Temperature dependence of Soret and diffusion coefficients for toluene-cyclohexane mixture measured in convection-free environment, J. Chem. Phys. 143 (22) (2015) 224902.
[40] V. Shevtsova, C. Santos, V. Sechenyh, J.C. Legros, A. Mialdun, Diffusion and soret in ternary mixtures. Preparation of the DCMIX2 experiment on the ISS, Microgravity Sci. Technol. 25 (2014) 275–283.
[41] T. Grossmann, J. Winkelmann, Ternary diffusion coefficients of cyclohexane + toluene + methanol by taylor dispersion measurements at 298.15 K. Part 1. Toluene-rich area, J. Chem. Eng. Data 54 (2) (2009) 405–410.
[42] T. Grossmann, J. Winkelmann, Ternary diffusion coefficients of cyclohexane + toluene + methanol by taylor dispersion measurements at 298.15 K. Part 2. Low toluene area near the binodal curve, J. Chem. Eng. Data 54 (2) (2009) 485–490.
[43] I.I. Ryzhkov, V.M. Shevtsova, On thermal diffusion and convection in multicomponent mixtures with application to the thermogravitational column, Phys. Fluids 19 (2) (2007) 027101.

[44] B. Seta, A. Errarte, A. Mialdun, I.I. Ryzhkov, M. Bou-Ali, V. Shevtsova, Cross-diffusion governs an oscillatory instability in a ternary mixture with the soret effect, Phys. Chem. Chem. Phys. 25 (2023) 15715–15728.
[45] B. Seta, A. Errarte, I.I. Ryzhkov, M. Bou-Ali, V. Shevtsova, Oscillatory instability caused by the interplay of Soret effect and cross-diffusion, Phys. Fluids 35 (2) (2023) 021702.
[46] S. Prokopev, T. Lyubimova, A. Mialdun, V. Shevtsova, A ternary mixture at the border of Soret separation stability, Phys. Chem. Chem. Phys. 23 (2021) 8466–8477.
[47] B. Seta, A. Errarte, D. Dubert, J. Gavalda, M.M. Bou-Ali, X. Ruiz, Gravitational stability analysis on double diffusion convection in ternary mixtures, Acta Astronaut. 160 (2019) 442–450.
[48] M. Larranaga, M.M. Bou-Ali, E. Lapeira, C. Santamaria, J.A. Madariaga, Effect of thermophyisical properties and morphology of the molecules on thermodiffusion coefficient of binary mixtures, Microgravity Sci. Technol. 26 (2014) 29–35.
[49] M.M. Bou-Ali, O. Ecenarro, J.A. Madariaga, C.M. Santamaría, J.J. Valencia, Stability of convection in a vertical binary fluid layer with an adverse density gradient, Phys. Rev. E 59 (1999) 1250–1252.
[50] A. Zebib, Stability of ternary and binary mixtures in a vertical slot including the soret effect, J. Chem. Phys. 129 (13) (2008) 134711.
[51] I.I. Ryzhkov, V.M. Shevtsova, Convective stability of multicomponent fluids in the thermogravitational column, Phys. Rev. E 79 (2009) 026308.
[52] T. Lyubimova, N. Lobov, V. Shevtsova, Stability of a stationary plane-parallel flow of a ternary fluid between two vertical plates maintained at constant different temperatures, Eur. Phys. J. E 41 (2018) 23.
[53] D. Bratsun, A. Mizev, E. Mosheva, Extended classification of the buoyancy-driven flows induced by a neutralization reaction in miscible fluids. Part 2. Theoretical study, J. Fluid Mech. 916 (2021) A23.
[54] E. Ruckenstein, Can phoretic motions be treated as interfacial tension gradient driven phenomena? J. Colloid Interface Sci. 83 (1) (1981) 77–81.
[55] J. Anderson, Colloid transport by interfacial forces, Annu. Rev. Fluid Mech. 21 (1) (1989) 61–99.
[56] S. Marbach, L. Bocquet, Osmosis, from molecular insights to large-scale applications, Chem. Soc. Rev. 48 (11) (2019) 3102–3144.
[57] S. Marbach, H. Yoshida, L. Bocquet, Local and global force balance for diffusiophoretic transport, J. Fluid Mech. 892 (2020) A6.
[58] J.F. Brady, Particle motion driven by solute gradients with application to autonomous motion: continuum and colloidal perspectives, J. Fluid Mech. 667 (2010) 216–259.
[59] S. Shin, Diffusiophoretic separation of colloids in microfluidic flows, Phys. Fluids 32 (10) (2020) 101302.
[60] R.R. Raj, C.W. Shields, A. Gupta, Two-dimensional diffusiophoretic colloidal banding: optimizing the spatial and temporal design of solute sinks and sources, Soft Matter 19 (5) (2023) 892–904.
[61] R. Golestanian, T.B. Liverpool, A. Ajdari, Propulsion of a molecular machine by asymmetric distribution of reaction products, Phys. Rev. Lett. 94 (22) (2005) 220801.
[62] R. Golestanian, T.B. Liverpool, A. Ajdari, Designing phoretic micro- and nano-swimmers, New J. Phys. 9 (5) (2007) 126.
[63] J.L. Moran, J.D. Posner, Phoretic self-propulsion, Annu. Rev. Fluid Mech. 49 (1) (2017) 511–540.
[64] R. Cerbino, A. Vailati, M. Giglio, Soret driven convection in a colloidal solution heated from above at very large solutal Rayleigh number, Phys. Rev. E 66 (5) (2002) 055301.
[65] A. Ryskin, H. Pleiner, Thermal convection in colloidal suspension with negative separation ratio, Phys. Rev. E 71 (2005) 056303.
[66] F. Winkel, S. Messlinger, W. Schöpf, I. Rehberg, M. Siebenbürger, M. Ballauff, Thermal convection in a thermosensitive colloidal suspension, New J. Phys. 12 (5) (2010) 053003.
[67] I.N. Cherepanov, B.L. Smorodin, Influence of sedimentation length on the convective stability of a colloidal suspension, J. Exp. Theor. Phys. 125 (6) (2017) 1199–1207.
[68] I.N. Cherepanov, B.L. Smorodin, Convection of strong stratified colloidal suspension, Microgravity Sci. Technol. 31 (5) (2019) 517–525.
[69] I. Cherepanov, B. Smorodin, Traveling waves of a colloidal suspension in a closed cell, Eur. Phys. J. E 45 (4) (2022).
[70] B.M. Alessio, A. Gupta, Diffusiophoresis-enhanced Turing patterns, Sci. Adv. 9 (45) (2023) eadj2457.
[71] P.K. Maini, T.E. Woolley, R.E. Baker, E.A. Gaffney, S.S. Lee, Turing's model for biological pattern formation and the robustness problem, Interface Focus 2 (4) (2012) 487–496.