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Additional transport by oscillatory buoyancy driven convection in diffusion experiments

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Abstract: A model for the transverse g-jitters induced transport in binary diffusion experiments is presented. The flow is supposed parallel and oscillating at a single frequency supposed high compared to the solute lateral diffusion time scale. This model applies to long aspect ratio experiments. A time-averaged effective diffusivity approach allows us to recover several recently published results and to provide a better understanding of the underlying physics. In the case of a given fully established velocity profile, the variation of the effective diffusion coefficient is such that $(D_{eff}-D)/D$ follows an ω^{-2} or an $\omega^{-7/2}$ law respectively for low and high frequency compared to the momentum lateral diffusion time scale. The transport by this type of flow is shown to be hardly significant in typical impurity diffusion experiment in microgravity.

1. INTRODUCTION

A number of recent works deals with the additional transport due to vibration induced flows in microgravity binary diffusion experiments. After his long-capillary space experiments, Smith [1,2] claims that this mode of transport can indeed lead to a significant overestimate of the measured coefficient: a ratio higher than two is observed on the PbAu system depending whether the MIM (Microgravity Insulation Mount) vibrations insulator facility was active or not. He also made one experiment with forced oscillations (4mg at 0.1Hz, oriented at 45° with respect to the capillary), which shows a strong enhancement of the transport coefficient. On the other hand, shear-cell experiments in the FOTON 12 mission [3] and ground-based measurements with electromagnetic damping of convection [4] show a very good agreement for the SnIn and SnBi systems. The problem is that the understanding of additional transport in space experiments is not yet deep enough on the experimental process (e.g. geometry, density ratio of the couple, experiments duration, etc.) and/or environment (e.g. residual g spectrum) to provide definite criteria on the possible influence of convection in a given microgravity experiment.

Mathiak and Willnecker [5] developed an original numerical method to investigate the transport by an oscillating flow in a diffusion experiment. The random walk of a particle is simulated in the cross section of a capillary tube of diameter H (our notation); given an arbitrary longitudinal velocity profile in the cross section, the axial convective transport of this particle is calculated at each time step depending on its lateral position at this step. Several velocity profiles of shape $u(r)=v_0(r)\sin(\omega t)$ were tested and the effective diffusion coefficient D_{eff} exhibited a clear $D_{eff}-D \propto v_0^2/H^2\omega^2$ behaviour at high frequency for all the realistic velocity profiles considered.

Compared to this work, the numerical simulation of oscillatory solutal convection in a diffusion experiment configuration by Matsumo and Yoda [6] has the interest of integrating the coupling between the composition and the velocity fields. They found an apparent diffusion coefficient independent of frequency at high frequency; with their parameters the diffusion coefficient is overestimated by about 8%.

However, as in their calculation, the gravity amplitude g is inversely proportional to its frequency over most of the investigated range, their results are compatible with a $(g/\omega)^n$ power law with some exponent n to be determined. Though few details are given on the comparison, Matsumo and Yoda observed a 6% difference only between 2D and 3D simulations of the concentration field.

Naumann [7] performs an analytical study of the 2D problem assuming a parallel flow in the cavity. He expects the discrepancy between 2D and 3D-cylindrical models to be minor. For a single periodic acceleration, his solution, under the form of a trigonometric series, yields a $Gr^2/Fq^{-7/2}$ behaviour of the transport at high Prandtl or Schmidt and high frequency. The Grashof number is $Gr=g(\beta_c\nabla c+\beta_T\nabla T)H^4/\nu^2$, where β_C and β_T are respectively the solutal and thermal expansion coefficient, • is the kinematic viscosity, and g the amplitude of the oscillating acceleration transverse to the cavity. Older papers [8,9] also relate such a dependence concerning very similar flows: they present a modified Taylor's dispersion approach of forced oscillatory pipe flows involved in environment and biomechanics problems.

The aim of the present paper is to present a phenomenological approach on additional mass transport due to an oscillating flow in a diffusion experiment. The presented order of magnitude approach allows one to recover the results of Naumann [7] and Mathiak and Willnecker [5] and emphasises the underlying physics. It is important to keep in mind that we will not consider two convective effects reported in the literature that could be of interest in mass transport problems in microgravity. The first one is the transport by the non solenoidal flow resulting from solutal expansion of the fluid, treated by Perrera and Sekerka [10]. The second one, more closely linked to the present work, is the transport by a non-zero time averaged flow induced by g-jitters, as studied by Gershuni [11] and Savino and Monti [12]. We voluntarily focus here on the case of non-stationary flows. The case of quasi-stationary flows can be treated through our model based on Taylor's approach [13].

2. CONFIGURATION, HYPOTHESIS AND TRANSPORT EQUATIONS

The configuration we work on is illustrated in figure 1. The capillary tube of a binary diffusion experiment is assimilated to a 2D cavity with impermeable walls. The ratio of its diameter H to its length

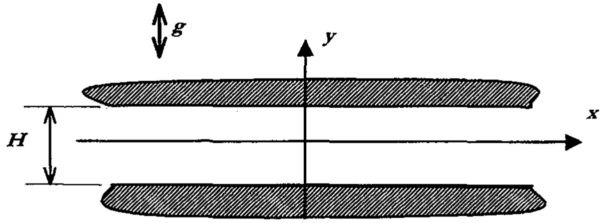

Figure 1 : Configuration of the diffusion experiment

L, denoted ε, is very small. The concentration field is split into its average over the cross section, $c_o(x,t)$ and its variation in the section, $c_1(x,y,t)$. In the case of a quasi-parallel flow $\mathbf{u}=u(x,y,t)$, and after a duration t from the beginning of diffusion, such that

$$t \gg H^2/D, \tag{1}$$

the solute transport can be described by [14]:

$$\frac{\partial c_0}{\partial t}+\frac{\partial}{\partial x}\langle uc_1\rangle = D\frac{\partial^2 c_0}{\partial x^2} \tag{2}$$

and

$$\frac{\partial c_1}{\partial t}+u\frac{\partial c_0}{\partial x} = D\nabla_S^2 c_1, \tag{3}$$

where <.> and ∇_S^2 are respectively the average and Laplacian operators over the cross section. Equation (3) holds provided that $c_1 << c_0$. In contrast with the usual Taylor dispersion approach the time derivative is conserved in (3), to allow the account of temporal fluctuations of c_1. In the case of a quasi steady flow, the time derivative in (3) can be neglected and our former model of effective diffusion [4,14] can be used, if the velocity field is known.

We focus all along the present paper on the effect of a high-frequency pulsating velocity field driven by a single frequency g-jitter. An additional hypothesis is that for the relatively weak flows expected in the microgravity environment, the non-linear terms in the Navier-Stokes equation can be safely neglected, meaning that the gravity and the g-jitter both have the same pulsation ω. Note that the high frequency hypothesis is defined relatively to radial diffusion time scale:

$$\omega H^2/D >> 1, \tag{4}$$

For typical experimental conditions, H = 1.5 mm and $D = 2\ 10^{-9}$ m²/s, the above equation implies $\omega >> 10^{-3}$ rad/s, a condition easily met in practice. As soon as the variations of c_0 spreads on a distance L_C greater than H, hypothesis (4) allows one to consider c_0 is unchanged during one period and one can work on the time averaged transport of the mean concentration. Equation (2) is then integrated over one period and becomes:

$$\frac{\partial c_0}{\partial t} + \frac{\partial}{\partial x}\left(\frac{\omega}{2\pi}\int_0^{\frac{2\pi}{\omega}} \langle uc_1 \rangle dt\right) = D\frac{\partial^2 c_0}{\partial x^2}. \tag{5}$$

The aim of the model is to express the $<uc_1>$ term as a function of $\partial c_0/\partial x$ to obtain a 1D diffusion equation for c_0. The effective time averaged diffusivity will then be given by:

$$D_{eff} = D - \frac{\left(\frac{\omega}{2\pi}\int_0^{\frac{2\pi}{\omega}} \langle uc_1 \rangle dt\right)}{\partial c_0/\partial x} \tag{6}$$

3. ORDERS OF MAGNITUDE ANALYSIS

3.1 No boundary layer flow

Following the work of Thevenard and Benhadid [15], we should keep in mind that, when the frequency of the g-jitters gets far higher compared to the radial transport of momentum time scale, the velocity field exhibits a "core and boundary layers" structure. As we are interested in high Schmidt number fluids (Sc~100 in liquid metals), hypothesis (4) is possibly compatible with either high or low frequency velocity field, from a hydrodynamic point of view. We focus first on the relatively low frequency case, were no singularity is present in the velocity field. Taking into account hypothesis (4), the range of validity of this assumption can be written as :

$$Fq = \omega H^2/\nu \leq 1 \text{ and } FqSc >> 1. \tag{7}$$

In equation (3), the orders of magnitude of the temporal and diffusive terms are:

$$D\nabla_S^2 c_1 \sim \frac{Dc_1}{H^2} \text{ and } \frac{\partial c_1}{\partial t} \sim \omega c_1.$$

To the first order, hypothesis (4) leads then to neglect the diffusive term in (3), which can be solved for c_1 to find:

$$c_1 \sim -\frac{u}{\omega}\frac{\partial c_0}{\partial x}. \tag{8}$$

The straightforward use of this expression in equation (2) would lead to a zero time averaged additionnal transport, since u and c_1 are in quadrature. This explains the low efficiency of oscillatory flows in terms of additional transport when time averaged equations are concerned. More generally, what happens is that the diffusive term in (3) introduces some phase shift between u and c_1 thus leading to some net transport by convection. To progress in the investigation of the time averaged transport property of the flow, equation (3) can be multiplied by c_1, in order to get the uc_1 product:

$$c_1\frac{\partial c_1}{\partial t} + uc_1\frac{\partial c_0}{\partial x} = D\, c_1\nabla_S^2 c_1. \tag{9}$$

In the time averaging operation, the first term disappears, and averaging over the cross section yields:

$$\langle uc_1\rangle = D\frac{\langle c_1\nabla_S^2 c_1\rangle}{\partial c_0/\partial x}. \tag{10}$$

The right hand side of (10) can be evaluated making use of (8), and the order of magnitude of the time averaged effective diffusivity is found with (6):

$$\langle uc_1\rangle \sim D\frac{u^2}{\omega^2 H^2}\frac{\partial c_0}{\partial x}, \text{ so that } \frac{D_{eff}-D}{D} \sim \frac{u^2}{\omega^2 H^2}. \tag{11}$$

We thus recover the scaling law initially proposed by Mathiak and Willnecker [5] from numerical results. As u is expected to be independent of ω at frequencies $Fq=\omega H^2/\nu<1$ and to follow a ω^{-1} law for higher frequencies, both ω^{-2} and ω^{-4} power laws can *a priori* be expected. However, in the high frequency case, a closer look to the different regions of the parameter space is necessary, as will now be developed.

3.2 Boundary layer flow

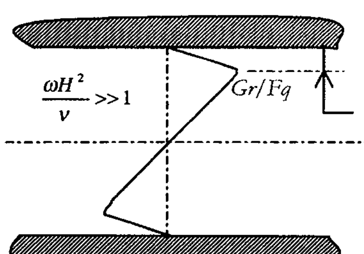

Figure 2 : Typical high frequency velocity profile

We now focus on the case of a parallel flow oscillating at high frequency $\omega H^2/\nu >> 1$, whose main features are represented on figure 2, in terms of velocity amplitude. A more exact description, as given in [15], requires the knowledge of the phase shift of the flow across the section, varying from zero at the walls towards $\pi/2$ in the middle of the cavity for infinitely high frequency. The velocity amplitude exhibits a linear core profile and two boundary layers at the walls to satisfy the no slip condition. The order of magnitude of the maximum amplitude is $u \sim \frac{\nu}{H}\frac{Gr}{Fq}$ and the thickness of the layers is $\delta \sim H/Fq^{1/2}$ [15]. As we have seen from equation (8) that c_1 follows the flow (except in solutal boundary layers far thinner – a factor Sc smaller - than the hydrodynamics ones), it has the same structure. It is still licit to use equation (10), but an evaluation of the relevant terms needs to be done in each region of the flow. In the inviscid core, $\nabla_S^2 c_1 = 0$, thus transport only occurs through the boundary layers. Consequently we have to estimate:

$$\left\langle c_1 \nabla_S^2 c_1 \right\rangle = \frac{2}{H} \int_0^\delta c_1 \nabla_S^2 c_1 \, dy .$$

In the layers, $c_1 \nabla_S^2 c_1 \sim \left(\frac{c_1}{\delta}\right)^2 \sim \left(\frac{u}{\omega\delta}\frac{\partial c_0}{\partial x}\right)^2$, and the average operator is $\frac{1}{H}\int_0^\delta dy \sim \delta / H \sim Fq^{-1/2}$, thus substituting to its value and:

$$\frac{D_{eff} - D}{\mathrm{D}} \sim \frac{Gr^2}{Fq^{7/2}}. \tag{12}$$

This last result allows one to explain Naumann's result for high Schmidt number and high frequency, as a transport through boundary layers. Note that the expected ω^{-4} behaviour expected from Mathiak and Willnecker [5] can hardly be observed in "real flows", since the occurrence of a ω^{-1} dependent velocity requires the boundary layer regime to be established.

4. ANALYTICAL SOLUTION BASED ON THEVENARD AND BENHADID VELOCITY PROFILE

Given an analytical form for the velocity profile, it is possible to follow step by step the calculations presented in section 3. The velocity profile derived by Thevenard and Ben Hadid [15] for a differentially heated 2D elongated cavity with monochromatic transverse g-jitters is a good candidate for this exercise. It is given by:

$$U(Y) = j\frac{Gr_T}{Fq}\left(Y - \frac{1}{2}\frac{sh(\sqrt{jFq}Y)}{sh(\sqrt{jFq/2})}\right)e^{j\omega t}. \tag{13}$$

The scalings used are ν/H for the velocity and H for lengths. The thermal Grashof number Gr_T is based solely on the temperature gradient. However, as diffusivity experiments as well as many type of microgravity experiments feature composition gradients, it is interesting to extend this approach to the case of solutal buoyancy driven convection. When hypothesis (1) is verified, axial composition variations occur on a typical length scale higher than the capillary diameter, and the composition gradient can be considered uniform in a first approximation. It is then possible to replace the temperature gradient in the Grashof number by $\partial c_0/\partial x = \Delta C_0 / H . \partial C_0 / \partial X$ (ΔC_0 being the height of the initial concentration step). Moreover, considering eqs. (1) and (4), it can be shown that the time variations of $\partial C_0/\partial X$ are small enough for eq. (13) to remain valid.

4.1 Constant composition gradient

We here assume that the velocity field consists in a single loop, which is so elongated that the flow is quasi parallel and quasi steady and we neglect the driving action of the gradient in concentration fluctuations $\partial c_1/\partial x$. This alternate velocity field will thus be written:

$$U(Y) = j\frac{Gr_S}{Fq}\frac{\partial C_0}{\partial X}\left(Y - \frac{1}{2}\frac{sh(\sqrt{jFq}Y)}{sh(\sqrt{jFq/2})}\right)e^{j\omega t} \quad \text{with} \quad Gr_S = \frac{g\beta\Delta C_0 H^3}{\nu^2}, \tag{14}$$

After tedious calculations, the time averaged transport equation (5) is found to become:

$$\frac{\partial C_0}{\partial \tau}=\frac{\partial}{\partial X}\left\{\left[1+S^2\left(\frac{\partial C_0}{\partial X}\right)^2\right]\frac{\partial C_0}{\partial X}\right\}$$

$$\text{where } S^2=\frac{Gr_S^2Fq^{-4}}{8}\left(\gamma\frac{\mathrm{sh}\gamma+\sin\gamma}{\mathrm{ch}\gamma-\cos\gamma}-2\right) \text{ and with } \gamma=\sqrt{\frac{Fq}{2}} \text{ and } \tau=Dt/H^2. \tag{15}$$

This equation is exactly analogous to the transport equation derived by Maclean and Alboussière [14] in the case of steady solutal convective transport in a diffusion experiment. As noticed in ref. [14], it can be re-scaled in order to get a universal equation, all the parameters being absorbed in the rescaling of τ and X. The non dimensional effective diffusivity, D_{eff}/D, is here the term in square brackets. One recovers the two asymptotic regimes predicted by the analysis of section 3:

- for high frequency, $$\frac{D_{eff}-D}{D}\approx\frac{Gr_S^2Fq^{-7/2}}{8\sqrt{2}}\left(\frac{\partial C_0}{\partial X}\right)^2, \tag{16}$$
- for low frequency, $$\frac{D_{eff}-D}{D}\approx\frac{Gr_S^2Fq^{-2}}{2880}\left(\frac{\partial C_0}{\partial X}\right)^2. \tag{17}$$

4.2 Time evolving composition gradient

For the case of diffusion coefficients measurement experiments, the convection driving force evolves continuously with the longitudinal diffusion scale, and it is necessary to account for this variation to make meaningful predictions. For instance, a rapid look at eq. (15) would seem to indicate that D_{eff} exhibits a S^2 dependence. However, Botton *et al.* [4,16] have shown that its true scaling law is D_{eff}~S. This can be shown by plugging the expression $\partial C_0/\partial X = H^2/D_{eff}t$ in equation (15).The above expression also allows one to derive the time variation $D_{eff}\sim t^{-1/2}$.

As shown in [16], an approximate solution of equation (15) can be found under the form $C_0(X,\tau)=f(X/\delta(\tau))$, where f is an arbitrarily chosen function. At the end of a diffusion experiment, it is actually what is done when the concentration profile is analysed assuming an error function of shape: $c_0(x,t)=\frac{\Delta C_0}{2}erfc(\frac{x}{2\sqrt{D_{app}t}})$, to obtain an apparent diffusion coefficient D_{app}. In this case, the solution given under implicit form by [16] is:

$$\frac{D_{app}}{D}=1+\frac{S^2}{4\pi\tau}\ln\left(1+\frac{4\pi\tau}{S^2}\frac{D_{app}}{D}\right). \tag{18}$$

This estimate of the apparent diffusion coefficient is plotted in figure 3 as a function of $4\pi\tau/S^2$. To estimate the value of $4\pi\tau/S^2$ in an *a priori* very sensitive space experiment, we can take:

$$H=2\ 10^{-3}\ m,\ g=10^{-3}\ m^2/s,\ \omega=10^{-2}\ rad/s,\ \beta=0.5\ (wt.fr)^{-1},\ \nu=10^{-7}m^2/s,\ \Delta C_0=10^{-1}wt.fr.$$

Then, Gr_S=40, Fq=0.4 and from (15) or its low frequency approximation, equation (17), S^2=3.47. A condition on τ is that it must be chosen in order to satisfy equation (1); τ=10 is then a minimum which yields $4\pi\tau/S^2$=36. Judging from figure 3, the overestimate of D is then of about 10%. Though this is not negligible, one must keep in mind that the parameters have been chosen to simulate a particularly critical experiment: usual Grashof numbers are often a factor ten smaller, leading to a hundred fold higher values of $4\pi\tau/S^2$ and consequently, the transport by oscillatory flow will be very weak.

For instance, in the experimental configuration of the Foton 12 experiment on Sn-In-Bi alloys, our model predicts that additional transport due to g-jitters can be safely neglected. Indeed, inserting relevant values $H=1.5\ 10^{-3}$ m, $g=10^{-3}$ m²/s, $\omega=10^{-2}$ rad/s, β=0.3 (wt.fr.$^{-1}$), ν= $3.10^{-7}m^2/s$, $\Delta C_0=2.5\ 10^{-2}$ wt.fr., we get $S^2 = 4.8\ 10^{-5}$. The model predictions should also be considered in connection with experimental results obtained in the Madylam laboratory through the application of a steady magnetic field to control fluid flow [4]. The agreement between FOTON 12 and ground based data is a clear indication that convective transport was actually negligible in the FOTON 12 experiment.

An apparent weakness of our present approach is that the straight forward application of these formulas to the case of Smith's experimental point with forced oscillations would lead to negligible convective transport. More complex phenomenon can actually be expected in Smith configuration, since the g-jitters is not transverse, but makes a 45° angle with the axis of the cavity. Note moreover that hypothesis (1) does not hold: even though the duration of this particular experiment is reported neither in [1] nor in [2], one can expect from reported duration of other experiments that it was of order 1 hour, whereas the radial diffusive time is about 6000 seconds.

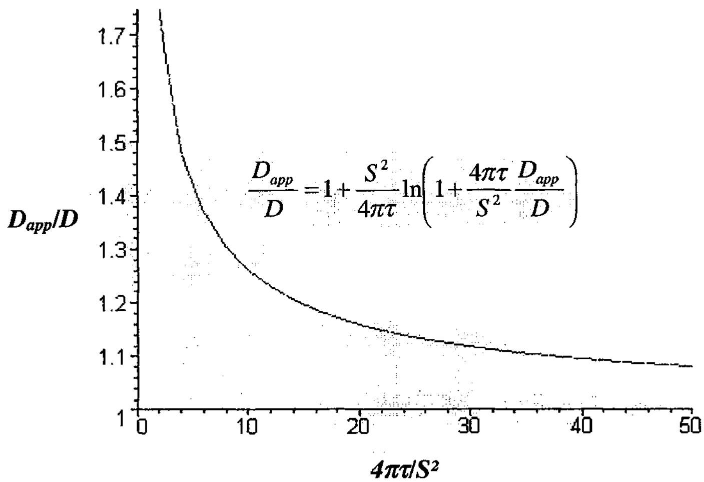

Figure 3 : Apparent diffusion coefficient estimated by equation (18).

5. CONCLUSION

Our purpose in this paper was to investigate the effect of g-jitters induced solute transport in diffusion coefficient measurement configurations. A simple model has been presented through both order of magnitude analysis and fully analytical approach. This modified Taylor's dispersion approach allows one to recover several recently published results and to provide a better understanding of the underlying physics. We consider only high frequency fluctuations, such that $\omega H^2/D >> 1$. In practice, oscillation pulsations in the range $\omega >> 10^{-3}$ rad/s can be considered as high frequency in terms of radial solute transport. The duration of the experiment is considered long enough to allow for the use of time averaged equations.

Due to the high values of the Schmidt numbers usually met in the experiments, these hypotheses are compatible with the existence of both low and high frequency hydrodynamic regimes. Order of magnitude arguments show that, in the low hydrodynamic frequency range and for a given fully established velocity profile, the effective diffusivity is such that $(D_{eff}-D)/D$ scales as ω^{-2}. In the high hydrodynamic frequency range, we showed that the existence of momentum boundary layers needs to be considered to recover Naumann's analytical results, $(D_{eff}-D)/D \propto \omega^{-7/2}$.

An approximate analytical solution of the solute conservation equations, based on a velocity profile initially derived by Thevenard and Ben Hadid, was also obtained. This analytical solution allows to derive an apparent diffusion coefficient as a function of the newly defined non-dimensional parameter S and to bridge both asymptotic regimes. In solutal buoyancy flows where the driving force decreases as $(D_{eff}t)^{1/2}$, $(D_{eff}-D)/D$ scales as respectively $\omega^{-1}/t^{1/2}$ and $\omega^{-7/4}/t^{1/2}$ in the low and high frequency ranges. Using this solution, we checked that the mass transport during the recent Foton 12 microgravity experiment was indeed diffusive. On the other hand, our results can not account for the experimental findings of Smith, since the orientation of the g-jitter in Smith's case may induce stronger flows than predicted by our model. Besides, the duration of his diffusion runs was too short for the modified Taylor's model to hold.

Future lines of work on the topic could thus include an extension of the effective diffusivity approach at small times and/or very low oscillation frequencies. Similarly, the amplitude of the time dependent oscillations of the effective diffusion coefficient around its mean value should also be investigated. Another important feature would be to check the range of validity of our oscillating flow solution, by comparing its predictions with those of thermovibrational models. However, the most important conclusion of the present work is that more well documented experimental data would be necessary to draw a definite conclusion on the topic.

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