MHD control of convection applied to chemical diffusivities measurements

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Résumé. La mesure au sol des coefficients de diffusion chimique dans les liquides est fortement perturbée par la convection naturelle. Dans le cas des métaux, nous avons étudié l'influence d'un champ magnétique capable de freiner considérabement la convection. Nous avons pu montrer expérimentalement sur le couple Sn-SnIn1%at qu'il est ainsi possible de retrouver la valeur obtenue en microgravité et que le freinage dû au champ magnétique est bien conforme aux prédictions théoriques.

Mots-clés : diffusion, champ magnétique, métal liquide, convection, étain.

Abstract. Chemical diffusivities measurements in liquids on the ground may be deeply influenced by buoyant convection. We have studied the influence of the braking effect of an uniform magnetic field on liquid metals. We have shown on the Sn-SnIn1at% couple that it was thus possible to measure a diffusion coefficient equal to that obtained in microgravity. Moreover, the braking effect of the magnetic field is found in agreement with theoretical predictions.

Key words : diffusion, magnetic field, liquid metal, convection, tin.

THE PRINCIPLE OF CHEMICAL DIFFUSIVITIES MEASUREMENTS : INFLUENCE OF CONVECTION

The principle of chemical diffusivities measurements is rather simple and has not basically changed since the first measurements performed at the end of the last century. Two elements of different composition are put into contact and interdiffusion occurs. After a given duration (between 10 hours in liquids, up to months in solids), the experiment is stopped, the concentration profile is analyzed and the diffusion coefficient is deduced from a fit between the experimental data and the purely diffusion theory. In the case of liquid metals, which is discussed in this paper, despite of the simplicity of the procedure, former reliable values of the diffusion coefficient are difficult to obtain and a scattering around 100% between different measurements is common. This discrepancy is usually attributed to convective effects in the liquid bulk. Because of the low value of the diffusion coefficient (about $10^{\circ}m^2s^{-1}$), even very low convective velocities (about 0.1 μ m/s) are able to enhance the transport of species and thus to modify the measured diffusion coefficient. Convection has two origins in those experiments :

- since measurements in liquid metals are made at temperatures above 300 °C, temperature differences can not be avoided and a thermal buoyancy force appears.
- the species involved do not have the same density and a solutal buoyancy force has also to be taken into account.

In both cases, the convective velocity depends on the Grashoff number [1]

 $Gr_i = \frac{\beta_i g G_i H^4}{v^2}$, where the subscribe *i* stands for t (thermal) or s (solutal), β the expansion coefficient, *g* the gravity, *G* the temperature or concentration

gradient, H the typical size of the experiment and v the kinematic viscosity.

Moreover, convective effects can not be detected on the experimental results. In the case of experiments made in long capillaries, the theoretical concentration profile is an error function, solution of the diffusion equation

$$\frac{\partial C}{\partial t} = D_0 \frac{\partial^2 C}{\partial x^2},\tag{1}$$

 D_o being exact the diffusion coefficient. It has been shown [2] that in the case of thermal convection, the average concentration in a cross section – which is the measured value - follows a similar law, but with an increased value of the diffusion coefficient :

$$\frac{\partial \langle C \rangle}{\partial t} = D_0 \left(1 + \alpha (Gr_t Sc)^2 \right) \frac{\partial^2 \langle C \rangle}{\partial x^2}, \qquad (2)$$

where $Sc = \frac{v}{D_0}$. Thus, the measured concentration profile is still an error function, but the fit with the experimental results will now give a diffusion coefficient enhanced by thermal convective transport.

Microgravity experiments do not suffer from this perturbation and the measurements made in these conditions are looked upon as a reference [3]. In ground based experiments, convection may be limited when reducing the capillary diameter (H^{t} dependence in the Grashoff number). However, for capillary diameter less than 1mm, experimental artifacts appear[4]. The vertical stabilizing configuration, where the heaviest element is at the bottom, has also been used. But small radial temperature gradients are difficult to avoid and the created convection can significantly disturb the measurement[5]. At last, in the

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case of liquid metals, an uniform magnetic field can damp significantly the movements in the liquid bulk. This is the approach that is presented here. We first briefly review the effect of a magnetic field in liquid metals. Then our experimental setup is described and preliminary results are presented. At least, we draw some general conclusions about the possibilities of magnetic fields.

HOW A MAGNETIC FIELD DAMPS CONVECTIVE EFFECTS

When a liquid metal flows through an uniform magnetic field \mathbf{B} , a current density \mathbf{j} appears given by the modified Ohm's law :

$$\mathbf{j} = \boldsymbol{\sigma} (\mathbf{E} + \mathbf{u} \times \mathbf{B}) \tag{3}$$

E being the electrical field and **u** the velocity. A Lorentz force $\mathbf{j} \times \mathbf{B}$ thus brakes the initial movement. The efficiency of MHD (Magnetohydrodynamic) braking depends essentially on the value of the electrical field . In the case of the classical Hartmann flow in a duct, if the walls are perfectly conducting, the current density **j** is short-circuited through the walls, the electrical field is zero and the braking follows a B⁻² law (($\mathbf{u} \times \mathbf{B} \times \mathbf{B}$). Now, if the walls are insulating, the electrical current loops have to close through the metal in thin layers. A strong electrical field is necessary and the braking follows a B⁻¹ law.

In the case of natural convection flows in capillaries (with insulating walls), the efficiency of the braking depends on the symmetry of the flow which allows the current loops to close within the fluid core without a strong electrical field. The most efficient configuration is an horizontal density gradient with a vertical magnetic field [6]. In this case, the MHD braking scales as B^{-2} . In a long capillary diffusion experiment in the case of thermal convection, the average concentration profile is governed by [2]

$$\frac{\partial \langle C \rangle}{\partial t} = D_0 \left(1 + \beta \frac{(Gr_i Sc)^2}{Ha^4} \right) \frac{\partial^2 \langle C \rangle}{\partial x^2}$$

where $Ha = \sqrt{\frac{\sigma}{\rho v}}BH$. Compared to expression(2), the convective perturbation

on the measurement is reduced by a factor varying as B^4 . The use of the magnetic field seems thus promising. However a possible drawback must be pointed out : the magnetic field (as a thermodynamic parameter) might modify also D_{α} , even if no clear experimental evidence for this can yet be given.

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THE EURIDICE EXPERIMENT. PRELIMINARY RESULTS;

The Euridice (Experimental Unit for Research In Diffusion. Influence of Convection Estimation) device has been specially designed to study the influence of an uniform magnetic field on the diffusion coefficient. It uses the now classical shear cell technique : the diffusion capillary is made of an alignment of cells (see figure 1). At the beginning, each capillary is filled with the liquid alloy (1a). Then the liquid metals are put into contact (1b). At the end, all the cells are turned, isolating small liquid elements, which are then cooled down and analyzed. This technique prevents segregations which could occur if the whole liquid sample was quenched. Alignment of the cells is controlled at each step of the experiment.



Figure 1 : Shear cell principle

The whole experiment is isolated from ground vibrations. Special attention has been given to the temperature control. Three heating elements (one compensating radial losses, the two other the axial losses along the capillary) ensure a good thermal homogeneity (less than 0.03°C along the 20 cm experiment at 300 °C). It is then possible to impose a perfect axial temperature gradient along the capillary by changing the regulation temperature of the two heating elements which control the axial losses. Notice that this approach is completely different from previous experiments where a minimal (but uncontrolled) convection level is searched by minimizing the capillary diameter, using a vertical stabilizing configuration. Here, on the contrary, a significant but now perfectly controlled convection is searched, so the influence of the magnetic field can be quantified. Our preliminary experiments were made with the Sn-SnIn1%at couple. The main reasons for this are the low melting point of the alloy which makes the heating technology easier and the very low density difference between tin and indium (perhaps the weakest in the periodic table, β . $= 6.10^{-1}$). In our experiments, a 68 K/m temperature gradient was applied. The

relative density variations due to temperature are thus 10^3 while those due to composition are 6.10^3 . Thus pure thermal convection is expected.

Different experiments with an applied magnetic field between 0.2 and 0.75 Tesla where performed. Figure 2 show two concentration profiles at different magnetic fields. : both curves can be properly fitted with an error function with a dispersion less than 3% (the error on the concentration measurement by chemical absorption). However, the two measured diffusion coefficients are completely different. This result shows that, as predicted, convective effects are undetectable on the experimental data.



Figure 2 : Two concentration profiles (left $D=2.4\ 10^{-9}\ m^2\ s^{-1}$, right 3.5 $10^{-9}\ m^2\ s^{-1}$) $D_{measured}$



Figure 3 : Variations of the measured diffusion coefficient with the applied magnetic field

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Figure 3 gives the variations of the measured diffusion coefficient with the magnetic field. It appears that the values follow a B^{-4} law with a low scattering (3%) as predicted by expression (4). It can thus be stated that the only effect of the magnetic field is to brake convective movements, not to modify D_0 . The asymptotic value is found to be $D_0 = 2.3 \cdot 10^{\circ} \text{m}^2 \text{s}^{-1}$, which is in fair agreement with previous microgravity results [3]. However, the results cut into two straight lines, an unpredicted result. This may be due to solutal effects. When the solutal density gradient has the same direction as the thermal one, the observed value is higher than when these gradients are opposite. It seems then that solutal density gradients, even when they are very low, may damp considerably the thermal convection movement.

CONCLUSIONS

These preliminary results show that the use of a magnetic field to improve chemical diffusion coefficient measurements in liquid metals is promising. Reproducible experiments where performed and the braking of convective effects was confirmed, as well as the B^{4} asymptotic law. The magnetic field in this range (less than 0.75T) does not modify the value of the diffusion coefficient. It has been possible to find a diffusion coefficient in the Sn-SnIn1at% couple similar to the one measured in microgravity (within the experimental scattering). But, the effect of composition buoyancy forces has to be precisely quantified, since it seems to modify the final result even when it is very weak.

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