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REVIEW

# Influence of Gravity on Atomic Mobility in a Liquid

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**Abstract**
Measurements of diffusion and thermodiffusion in liquids are very sensitive to convection caused for example by buoyancy. To reduce the impact of buoyancy-driven convection, benchmark experiments are performed in microgravity conditions. Here, we discuss the general influence of gravity on atomic mobility. The gravitational Péclet number and the gravitational length can be used to assess this influence. They show that the diffusion processes of atoms in a liquid is not affected by Earth's gravitational force but that the process is dominated by the thermal energy of the atoms. Data from experiments under different gravity conditions ranging from $10^{-5}g$ to $10^{6}g$ are summarized. They confirm that interdiffusion is only influenced by accelerations that are orders of magnitude larger than Earth's gravity.

**Keywords** Atomic mobility · Microgravity · Hypergravity · Diffusion · Metallic melt

## Introduction

In experimental measurements of interdiffusion using the so-called long-capillary technique, two samples of different composition are brought into contact to analyze the concentration distribution after a certain time. Interdiffusion describes the flux of particles induced by a gradient in the chemical potential due to a concentration difference. It influences the velocity of chemical reactions, solidification of multicomponent materials, sintering, and other processes (Cussler 2009; Grasso et al. 2020; Steinbach 2013). Measurements in solids can be performed in any orientation but in the case of liquid samples the denser liquid has to be placed at the bottom to stabilize the initial interface between the two compositions. Otherwise the samples will be mixed quickly due to buoyancy-driven convection (Kargl et al. 2013). Horizontal orientation would lead to an initially vertical interface that is unstable and would lead to immediate convection. Convection can also be provoked by free surfaces due to bubbles (Marangoni convection) (Ruiz and Pallarés 2011; Roşu-Pflumm et al. 2009) which is independent of gravity. Thus, measurements of interdiffusion in liquids can easily be disturbed by convection. To obtain benchmark values without buoyancy-driven convection, selected interdiffusion experiments were performed under microgravity (Mathiak et al. 1996; Garandet et al. (2004). The measured interdiffusion coefficient in microgravity was sometimes lower than the measured value obtained on ground (Praizey et al. 2001).

Denser particles in a liquid sediment over time due to gravity. This process can be accelerated by centrifugation which is used for example for blood fractionation. Sedimentation effects are known from the incongruent solidification of alloys, where solid parts nucleate with a different chemical composition than that of the melt, and thus rise or sink in the liquid depending on their relative density (Browne et al. 2017; Nguyen-Thi et al. 2011, 2014). Centrifugation of partially liquid alloys was also proposed as a method to purify scrap metal (Zhao et al. 2010).

In other words, the mass flow in liquids can be strongly influenced by gravity. In light of the fact that experiments in microgravity conditions do not always yield values that are in agreement with ground-based experiments, one might wonder whether gravity might influence diffusion

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processes by pulling the heavier atoms towards the bottom. As we point out in the following, this is in fact not the case. To see this, we will describe relevant parameters and critically assess diffusion experiments under different gravity conditions.

## Gravitational Péclet Number

For colloidal suspensions, where the diffusing particle is much bigger than the molecules or atoms comprising the surrounding fluid, a gravitational Péclet number $Pe_{g,c}$ is used to characterize the movement of the particles (Bérut et al. 2019; Russel et al. 1989). It is defined by the ratio of the particle weight and the Brownian thermal forces:

$$Pe_{g,c} = \frac{mgd}{k_B T} \tag{1}$$

with $d$ the diameter of the particles, $m$ its buoyant mass, $g$ the gravitational acceleration, $T$ the temperature of the system and $k_B$ the Boltzmann constant. The buoyant mass $m$ is the volume of the particle multiplied with the difference of density between the particle and the surrounding fluid.

As an example, silica particles with diameter $d \simeq 1\ \mu$m which are suspended in water have the gravitational Péclet number $Pe_{g,c} \simeq 1$. This means that particles will sediment quickly but also show random fluctuations induced by thermal agitation (Bérut et al. 2019). For colloidal particles in a density matched liquid $Pe_{g,c}$ would be well below unity. In this case sedimentation would not be completely absent but very slow. As another case, let us consider unagitated granular particles suspended in air. We then obtain $Pe_{g,c} \gg 1$ in agreement with the observation that the diffusive motion of unagitated granular particles is completely dominated by gravitational settling.

The Péclet number can also be written in terms of the gravitational length that characterizes the gravity-induced density dependence known from the barometric height distribution (Royall et al. 2005),

$$\ell_g = \frac{k_B T}{mg}, \tag{2}$$

with $Pe_{g,c} = d/\ell_g$. Gravitational settlement is dominant if the gravitational length $\ell_g$ is small compared to the size of the experimental sample. For the example of the silica particle mentioned above, $\ell_g \approx d$, and thus a noticeable density dependence is observed on the length scales relevant for the particle motion. Density matching in colloidal suspensions increases $\ell_g$ sufficiently far such that over the size of the experimental sample, gravitational settling becomes unimportant.

Applying this concept to atoms in a liquid alloy, we take as an example liquid zinc at 1000 K. Here the gravitational length is about $1.3 \times 10^4$ m. Note that we are here implying that thermal energy is the dominant driving factor for diffusive phenomena. Direct interatomic forces are much stronger, but due to Newton's third law, they mostly cancel out, leaving thermal fluctuations as the main driving force of mass transport. The gravitational length has to be compared to the height of the container. In a typical diffusion experiment the height of the container is in the order of $10^{-2}$ m which is orders of magnitude smaller than the gravitational length. This comparison shows that the influence of gravity on atoms in a liquid alloy can be neglected under normal and reduced gravity conditions.

The above estimates are based on a static energy balance. One might however wonder about dynamical effects. In fact, $Pe_{g,c}$ can be identified as the ratio of the time scale $t_D$ relevant for diffusion versus that relevant for sedimentation, $t_g$, identifying the colloidal regime as that where particles diffuse over the time window of experimental interest. This ascertains that for example, a brick in outer space, which formally obeys $Pe_{g,c} = 0$, is not dominated by diffusion on any reasonable time scale (Frenkel 2002). The time scale $t_D$ for diffusion is estimated from the time where the mean-squared displacement, $r^2 \sim Dt$, reaches the square of the size of the object itself, thus $t_D \sim d^2/D$. The time $t_g$ needed for a colloidal particle to be displaced by its own size under the influence of gravity is given by its sedimentation velocity $v_g \sim \mu m g$, where $\mu = D/k_B T$ is its mobility (and $m$ the buoyant mass). Hence, $t_g \sim d/\mu m g$, and setting $Pe_{g,c} = t_D/t_g$ recovers Eq. (1). For atomic systems or granular particles, where inertia effects play a role, $t_g$ is estimated from Newton's equation of motion as $t_g \sim \sqrt{d/g}$. The dimensionless number suitable to quantify the influence is then the gravitational Péclet number,

$$Pe_g = \frac{t_D}{t_g} = \frac{\sqrt{g} d^{3/2}}{D}. \tag{3}$$

The ratio is taken such that as gravitational effects become negligible, $Pe_g \ll 1$ (as then the typical time $t_g$ it takes for gravity to become noticeable, is much larger than the diffusion time), whereas $Pe_g \gg 1$ signifies strong gravitational effects. Related quantities known from fluid dynamics and granular matter physics are the Bond or Eötvös number (Clift and Grace 1978) quantifying the strength of gravitational forces over surface-tension forces, and the granular Bond number (Capece et al. 2015) estimating the relative importance of gravitational over cohesive forces. For liquid alloys typical values are in the order of $d \sim 1$ Å and $D \sim 10^{-8}\ \mathrm{m^2 s^{-1}}$.

The gravitational Péclet number is then in the order of $Pe_g \simeq 10^{-6}$ which is far below unity. By all accounts, liquid-state diffusion in alloys should be unaffected by Earth's gravity.

## Experiments Under Different Gravity Conditions

In order to suppress buoyancy-induced convection or to study the influence of gravity, some diffusion experiments were conducted under different gravity conditions. This includes mainly diffusion experiments in liquid samples performed under microgravity ($\mu$g) and normal gravity (1g) but also experiments under hypergravity.

Under gravity diffusion measurements can be disturbed by buoyancy e.g. due to inhomogeneous temperature distribution (Alexander et al. 1996). Measurements of self-diffusion in liquid pure metals sometimes lead to diffusion coefficients that were too high due to additional mass transport by convection. In this case, measurements under microgravity which where not disturbed by this effect where lower than the values obtained on ground (Kaptay 2008). This is especially relevant at temperatures well above the melting point. On the other hand, stable density layering in interdiffusion setups can dampen convection under gravity (Barat and Garandet 1996; Suzuki et al. 2005). On some microgravity platforms, e.g. parabolic flights, changes in the residual acceleration ("g-jitter") can lead to additional mass transport in liquid samples (Mathiak et al. 2005). Specially designed isolators which decouple the experimental setup from the structure of the platform reduce this influence (Smith et al. 2009). Marangoni convection caused by differences in surface tension due to bubbles or free surfaces can occur both on earth and in microgravity (Ruiz et al. 2015; Ruiz and Pallarés 2011; Roşu-Pflumm et al. 2009; Kargl et al. 2013).

Over the last decades experimental setups for diffusion measurements were optimized to reduce disturbing convection. For experiments where a stable density layering is possible and special care is taken to avoid bubbles and to minimize radial temperature gradients, measurements on ground and in micro-gravity show comparable results. Interdiffusion measurements in aluminum-based liquid alloys on ground and in micro-gravity agree within uncertainties (Sondermann et al. 2016, 2019; Garandet et al. 2004). In these experiments the shear cell technique was used where the two parts of the diffusion couple are melted separately and brought into contact after the annealing temperature has been reached. The concentration distribution along the diffusion couple is either recorded continuously by in situe X-radiography or at the end of the diffusion time by separating the liquid sample in several small parts and chemically analysing the solidified parts. The same technique was used on earth and in micro-gravity. The uncertainty of these interdiffusion measurements is about 10%. Also for Sn-Bi alloys it could be shown that experiments on ground give the same results as experiments done aboard a satellite (Roşu-Pflumm et al. 2009; Garandet et al. 2004; Praizey et al. 2001). Again the shear cell technique was employed.

A process that depends on diffusion in the melt is crystal growth. It has been reported that crystals of sodium chloride grow slower in microgravity compared to normal gravity conditions. This can be attributed to convective flow that transports depleted solution away from the crystal under normal gravity (Maruyama et al. 2002). Crystallization and crystal growth as well as the long-time dynamics in solidifying colloidal suspensions are in fact also known to be strongly influenced by gravity (Zhu et al. 1997; Simeonova and Kegel 2004).

Similar experiments that also probe the movement of atoms or molecules in a liquid are measurements of thermodiffusion. Thermodiffusion (also known as the Ludwig-Soret effect) describes the formation of a concentration gradient induced by a temperature gradient in a mixture. Comprehensive measurements of diffusion and thermodiffusion coefficients in water-isopropanol mixtures using three different instrumental techniques show good agreement between ground-based and microgravity experiments (Mialdun et al. 2012). Comparison between measurements of thermodiffusion in hydrocarbon mixtures onboard the International Space Station (ISS) and measurements on ground shows only minor variation in the values of the thermodiffusion coefficient (Ahadi and Saghir 2016; Larrañaga et al. 2015). However, in these studies not only the environment (microgravity and Earth gravity condition) was changed but also the experimental techniques. An overview over experiments on thermodiffusion in microgravity by the European Space Agency can be found here (Braibanti et al. 2019).

To further test the influence of gravity on single atoms, let us also take a look at measurements under hypergravity. Hypergravity means accelerations which are higher then the acceleration of Earth's gravity g. Ono et al. studied the miscible alloy In-Pb at about 140 °C, which is below its melting point, in a centrifuge that provides an acceleration of $10^6$g. They find that the Pb-content continuously increases in the direction of gravitational field from about 12 to 36 at.% Pb over the sample thickness of about 1.5 mm. This graded structure is continuous in atomic scale, and was formed by the sedimentation of solute atoms. The composition change reached a steady state within 60 hours (Ono et al. 2005). Taking as atomic radius $d \approx 10^{-10}$ m and the interdiffusion coefficient $D \approx 10^{-12}$ m$^2$/s (Ono et al. 2005) the gravitational Péclet number in this case is $Pe_g \simeq 3$. This is in accordance with the observation that hypergravity influences the movement of atoms. Alternatively, we can use Eq. 2 to calculate the gravitational length. At an acceleration of $10^6$g the gravitational length of lead at 140 °C is about $1.7 \times 10^{-3}$ m which is comparable to the thickness of the sample. A theoretical description to calculate the chemical distribution in a multicomponent system depending on the acceleration field is given by Mashimo (1994).

Similarly using a centrifuge, Ogata et al. compared measurements of the interdiffusion couple Cu-$Cu_{70}Zn_{30}$ at 400 °C in hyper-gravity of $10^6$g and in normal gravity of 1g. Depending on the direction of the concentration gradient with respect to the acceleration, the measured interdiffusion coefficient is 1.8 times larger than that of the 1g sample or only 0.83 times the value of the 1g sample (Ogata et al. 2015). These experiments show that gravity can lead to sedimentation of individual atoms even in solids. However, this effect is only measurable at extreme accelerations.

## Conclusion

In summary, we described the gravitational Péclet number and the gravitational length as parameters to assess the influence of gravity on individual atoms. Under normal conditions, the thermal energy in a liquid is larger than the gravitational potential energy by orders of magnitude. Thus by going from 1g to $\mu$g and in the absents of convection the atomic mobility itself is not affected. Only accelerations that are orders of magnitude larger than Earth's gravity influence interdiffusion.

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