The Manipulation of Paramagnetic Ions by Magnetic Field Gradient Forces

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Abstract

In this work the influence of the field gradient force on the distribution of dissolved paramagnetic ions is investigated. The experiments are performed with different concentrations of Fe^{2+}, directions of the field gradient and magnitudes of the magnetic induction. The dependencies derived in this project shall allow an optimized design of external magnetic fields to modify material properties by manipulating the distribution of ions in a fluid. An area of application is the controlled production of graded glasses or glass ceramics, from melts or other liquids in an external magnetic field.

Introduction

The effect of magnetic forces, acting on ions dissolved in aqueous liquids, has been discussed intensively in the recent years. Especially the capability of Lorentz forces to stir and mix liquids (glass melts, electrolytes) has been investigated [1,2]. In present works first applications for the adaption of magnetic equipment to feeders in glass production are tested [3].

Beside the Lorentz force, which appears only in combination of magnetic fields and electric fields, other magnetic forces show great promise for influencing material properties during the process of production. In presence of non-uniform magnetic fields and absence of electric fields the only appearing strong magnetic force which interacts with the magnetic moments of the ions of a solution is the magnetic gradient force (Kelvin force), which can be written in form of a force density as shown in (1).

$$f_{\nabla B} = \frac{\chi}{\mu_0}(B \cdot \nabla)B .$$

$$F_P = F_{\nabla c} + F_{\nabla H} = \frac{1}{2}\mu\chi_{mol}H^2\nabla c + \frac{1}{2}\mu\chi_{mol}(\nabla H)H$$

$$(1)$$

In this equation μ_0 means the permeability, χ susceptibility of the solution and B the magnetic flux density of the applied field. This force shows high potential for the use in material science and technology, as it is supposed to reach high magnitudes [4].

If a non-uniform magnetic field exists and paramagnetic ions with an effective magnetic moment are present (e.g. Mn^{3+}, Fe^{3+}, Fe^{2+}, Co^{2+}) [5], then the magnetic gradient force acts on this ions, depending on the magnitude of their magnetic moments and of the vector gradient. These relations are schematically shown for a Fe^{2+} - ion with $\mu_{eff} \sim 4.9 \ \mu_B$ ($\mu_B \sim 9.274*10^{-24}$ Am^2) and a flux density gradient in z - direction in Fig. 1.

In the present work the influence of the magnetic gradient force on the distribution of paramagnetic Fe^{2+} - ions in aqueous solutions is fundamentally investigated.
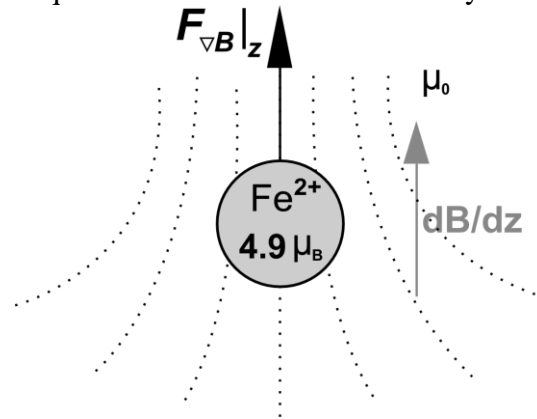

Fig. 1. Schematic of a paramagnetic ion influenced by the magnetic gradient force

The long-term objective is to transfer the information received from the experiments on aqueous solutions to real glass melts [6].

1. Experimental Procedure

To investigate the fundamental behaviour of liquids containing paramagnetic ions "cold" experiments at 25 °C were conducted. The used model liquids consist of p.a. grade Iron(II)chloride-tetrahydrate ($FeCl_2 \cdot 4H_2O$) dissolved in 3% sulphuric acid (H_2SO_4). The Fe^{2+} - concentrations of these solutions were measured with potentiometric titration before the experiments.

To apply the necessary magnetic field a cryogen-free superconducting DC magnet system (Cryogenics Inc.) with a maximum magnetic flux density B_{max} of 5 T was used. The cylindrical working chamber of this magnet system has a diameter of 300 mm and a depth of 400 mm.

In preliminary measurements the magnetic flux density B at different positions within the working chamber of the magnet system was measured. Based on the measured values the distribution of $B = f(r,z)$ and the magnetic vector gradient $(B \cdot \nabla)B = f(r,z)$ were estimated by numerical simulation [7]. The results are shown in Fig. 2.
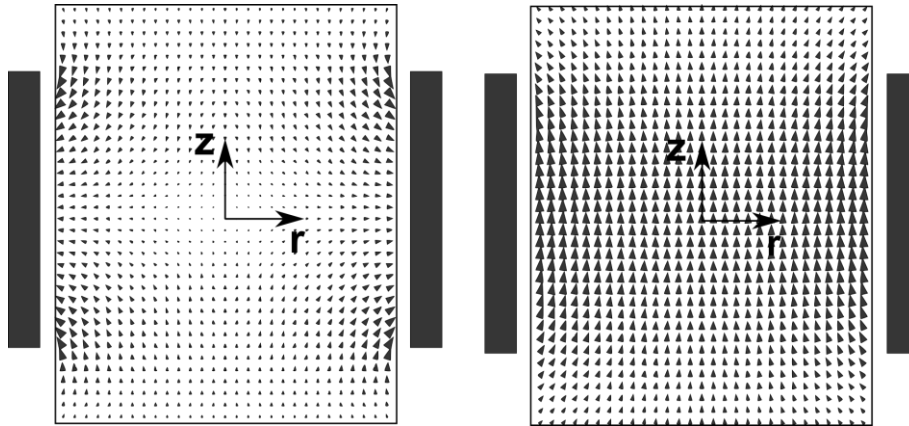
Fig. 2. Distribution of the vector gradient $(\boldsymbol{B} \cdot \nabla)\boldsymbol{B} = f(r,z)$ (left side) and distribution of them flux density $\boldsymbol{B} = f(r,z)$ (right side) within the working chamber of the cryogen-free magnet system

Future "hot" experiments with glass melts are planned with a high temperature furnace (XERION Advanced Heating Ofentechnik GmbH), which can be installed into the working chamber of the magnet system.

For the comparability of the experiments the positions of the measuring cell at which the "cold" experiments with model liquids were performed were chosen to fit the conditions of the "hot" experiments (z- and r-position ranges of the measuring cell: z = -180 mm up to +180 mm, r = 0 mm).

A cross-section of the magnet system and the cylindrical working chamber with the position of the measuring cell on position z = + 109 mm, r = 0 are shown in Fig. 3. The picture also shows the distribution of the z − component of the gradient force density $(\boldsymbol{B} \cdot \nabla)\boldsymbol{B}\,|_z / \mu_0 = f(r = 0,z)$, obtained by numerical simulations. At the position r = 0 and z = 109 mm the calculated gradient force has its maximum with approx. + 60 N/cm^3.
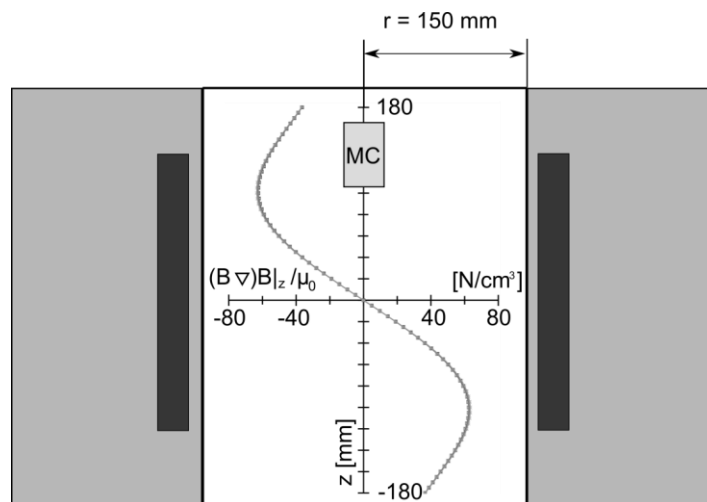

Fig. 3. Schematic cross-section of the cryogen-free magnet and the distributions of the magnetic gradient force density along the z-axis of the cylindrical working chamber (the position of the measuring cell is marked as "MC")

The measuring cell for the model liquids is double-walled, Fig. 4. This cell is tempered at 25 °C during the whole experiments to minimize influences from natural convection.
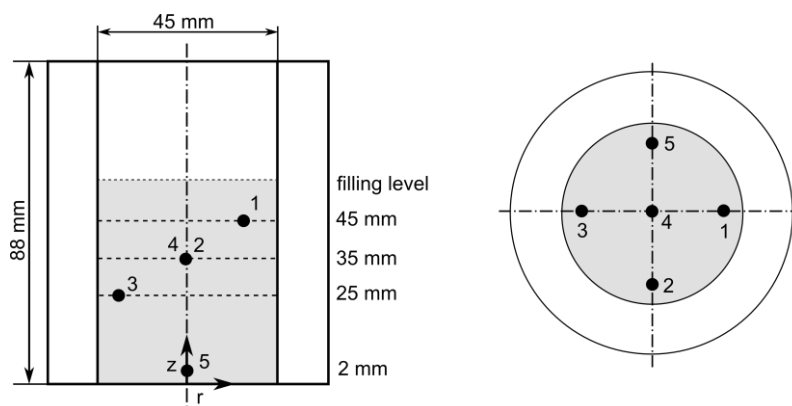

Fig. 4. Cross-section of the double-walled measuring cell, the black dots represent the positions at which the samples were taken.

The experiments itself were carried out for 12 hours with and without influence of the magnetic gradient field. After each experiment samples of 1 ml were taken at the positions shown in Fig. 4. These samples were analyzed by potentiometric titration to get the concentration of Fe^{2+} at the different sampling positions.

2. Results and Discussion

The results of an experiment carried out for 12 h with no magnetic gradient field applied are shown in Fig. 5 (triangles). Also displayed in this figure are the statistical values of measurements (grey lines for the arithmetical mean and upper/ lower confidence level) which were obtained outside the experimental setup prior to the experiment. As expected no change in concentration is noticeable if no gradient field is applied in the measuring cell. If a magnetic gradient field is active during the experiment a significant increase in the Fe^{2+} concentration is detectable (Fig. 5 dots). The highest concentration values are reached near the surface of the liquid.
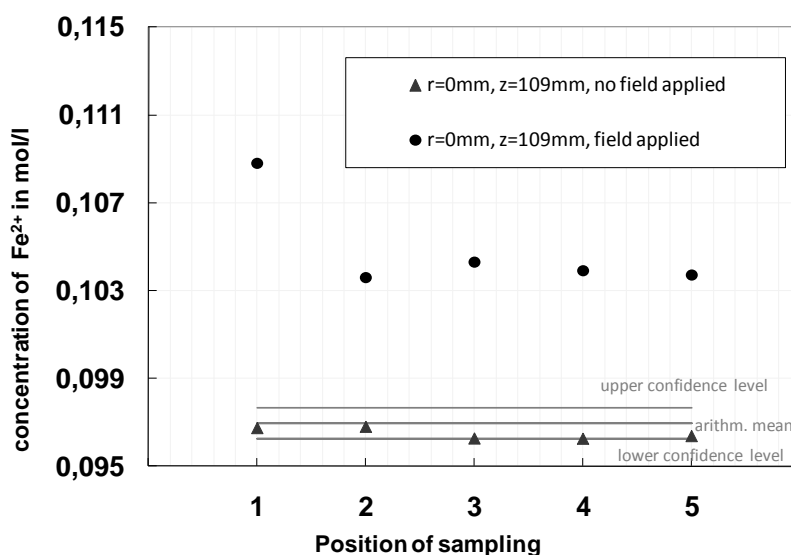
Fig. 5. Measurements of the Fe^{2+} concentration after the experiments conducted with no field applied (triangles). The detected concentrations after the experiments under influence of an applied field are shown as dots. The grey lines represent the results of statistical measurements prior to the experiments.

Conclusions

The fundamental usability of magnetic gradient forces acting on paramagnetic ions in an aqueous solution was proved. The experiments on model liquids consisting of paramagnetic ions (Fe^{2+}) showed a significant raise in the Fe^{2+} concentration if a magnetic gradient field is applied during the experiments. The next steps are experiments with lower and higher Fe^{2+} concentrations, with other positions of the measuring cell as well as the investigation of the dependence on the gradient force density.

Further investigations will include experiments on real glasses containing Fe^{2+} and Fe^{3+} as paramagnetic ions. Based on these results a model, which describes the interaction between gradient force distribution and the paramagnetic ions as well as its movements, will be created and verified with the experimental data.

Acknowledgement

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