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Invited paper

Microgravity research as a tool for the investigation of effects in magnetic fluids

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Abstract

The magnetic control of ferrofluids, provided by a magnetic volume force acting on the fluid in concurrence to gravitational forces, opens interesting fields for basic as well as application oriented research and for the design of applications using ferrofluids as acting media. The concurrence between magnetic and gravitational force respectively can often, especially in the investigation of new effects in not optimized systems, become a problem for experimental research with magnetic liquids. The possibilities to exclude gravitational forces by using special experimental facilities providing an environment with strongly reduced gravitational acceleration will be discussed, and the special chances for magnetic fluid research will be outlined here. Using a special example – the search for viscoelastic properties in commercial magnetic fluids – the interaction of usual terrestrial and microgravity experiments will be demonstrated. \bigcirc 1999 Published by Elsevier Science B.V. All rights reserved.

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1. Introduction

The magnetic control of properties and flows in magnetic fluids enables the creation and investigation of numerous interesting and technological relevant effects (see e.g. Refs. [1–3]). In experiments and theoretical investigations it is often found, that the magnetically induced effects are strongly coupled with gravitationally driven phenomena. This coupling can give rise to serious problems in

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the investigation of the magnetic effects. On the one hand it can enhance the problems in theoretical description of magnetic effects if complex coupled flow profiles have to be considered. Therefore the action of gravity is often neglected in theoretical approaches, making the comparison between laboratory experiments and theory difficult. On the other hand the effects driven by gravitational acceleration can be much stronger than the effects driven by the magnetic properties of ferrofluids. Thus the coupling may partly or completely suppress the magnetic effects or at least it may modify their appearance in experimental investigations.

A classical example for suppression and modification of magnetically driven effects in ferrofluids

by gravitational action is the normal field instability [4]. As Cowley and Rosensweig have shown, the critical magnetization, at which the instability occurs scales with $g^{1/4}$ (g gravitational acceleration), and the critical wavelength of the instability varies with gravitational acceleration like $q^{-1/2}$. This example shows on the one hand how gravity can suppress magnetically driven effects - reduction of gravitational acceleration will reduce the necessary magnetization of the fluid, making it easier to create the necessary magnetic fields driving the instability. On the other hand the change in the mutual strength' of the effects defining the wavelength of the instability - magnetic force, surface tension and gravitational acceleration - changes the qualitative structure of the effect expressed by the variation of the wavelength.

The example of normal field instability shows clearly, that it may be interesting for certain questions to reduce the gravitational forces acting on the experiment. Cowley and Rosensweig did this by means of a plateau experiment, i.e. by reduction of the density difference between the ferrofluid and the surrounding medium [4]. The disadvantage of such plateau experiments is given by the fact, that not only gravitational forces are changed, but also other parameters may vary if the ferrofluid is surrounded by a liquid of similar density. E.g. the surface tension of a ferrofluid-gas interface can be remarkably different from that of an interface between ferrofluid and another liquid. In addition one has to face the fact, that chemical processes between the ferrofluid and the surrounding liquid can give rise to an uncontrolled, irreversible change of the properties of the ferrofluid due to chemical processes.

The use of experiments under reduced gravity conditions – so-called microgravity experiments – can help to isolate the magnetic effects from gravitational influences and to make their quantitative investigation possible without disturbances from a surrounding liquid like it is used in plateau experiments. In addition the combination of usual laboratory experiments with microgravity research may give complementary results giving new insight into magnetic effects in ferrofluids. The challenges of microgravity experiments for research with magnetic fluids have already been shown in some first investigations concerning thermomagnetic convection, where normal buoyancy driven convection has been excluded to make the investigation of the pure thermomagnetic effect possible [5,6]. Further microgravity experiments on special phenomena in thermomagnetic convection have been proposed [7].

Nevertheless, beside all possible advantages, one should always face the fact that not only the magnetic effects in focus of a certain research question will experience an enhanced relative strength. Moreover all kind of disturbances, like wetting or effects due to inhomogeneities of the applied magnetic fields will also grow in importance. This became obvious in experiments on normal field instability [8], where a remarkable deviation from Rosensweigs theory was discussed, which could later on simply be related to inhomogeneities in the applied magnetic field. A simple demonstration experiment with a well defined homogeneous field [9] showed results exactly corresponding to the theory in Ref. [4].

In the following I will first introduce some experimental facilities providing an environment of strongly reduced gravitational acceleration, like they are commonly used in experiments. Afterwards the challenges of combinations between common terrestrial experiments and experiments under microgravity conditions will be illustrated with an example from the investigation of viscoelastic properties in ferrofluids.

2. Microgravity flight facilities

Beside the well known longterm flight opportunities using a space shuttle, sounding rockets or in near future the international space station, some earth bound facilities exist, providing fast and cheap access to a microgravity environment. In particular drop towers, drop tubes and parabolic flights provide a microgravity environment with residual acceleration below $10^{-2}g_0$ ($g_0 = 9.81$ m/s² normal gravitational acceleration) and with experimental times in the order between some seconds and some tens of seconds. The advantage of these flight facilities is on the one hand their comparably low price and on the other hand the easy access coupled with the possibility to use usual laboratory equipment for the performance of the experiments. The latter point is of high importance for the combination of terrestrial experiments with investigation in a microgravity environment. The use of an identical experimental setup for both kind of experiments makes the condition "microgravity" just an additional parameter that can be changed in the investigations. For the performance of the experiments described later on, we have used the drop tower in Bremen as well as parabolic flights.

In drop tower experiments the experimental setup is enclosed in a vacuum tight container which provides an internal power supply and a remote control and data link to the ground station. The container - usually called drop capsule - is dropped in an evacuated steel tube, which - in the case of drop tower 'Bremen' – provides a free fall height of 120 m corresponding to a free fall time of 4.75 s. Since the tube is evacuated, the capsule experiences no drag and thus falls freely in the earth's gravitational field. Therefore the experiment inside the capsule experiences a residual acceleration of only $10^{-6}g_0$. This residual acceleration is the best microgravity quality available nowadays. At the bottom of the tube the capsule is decelerated in a container filled with small polystyrene spheres. The maximum deceleration is in the order of $50q_0$ defining the stability limits for the experimental hardware. For more information on drop tower 'Bremen' see e.g. Ref. [10].

If longer experimental times under microgravity conditions are required, or if an investigation needs numerous experiments, e.g. for the change of several parameters, it may be preferable to use parabolic flights to perform the studies under microgravity conditions. In this case the hardware of several experiments is mounted in an airplane - the European Space Agency ESA uses an Airbus A300 – using power supply from the plane but being independent in all other belongings like data acquisition etc. The plane follows a parabolic trajectory starting from a normal flight level in about 9000 m, being pulled up to about 10 500 m with an inclination angle of about 45° at the begin of the parabola. Then the plane is guided into the free fall part of the trajectory where the thrust is only used

to compensate the remaining air drag, reducing the residual acceleration in the plane to $10^{-2}-10^{-3} g_0$. At the end of the free fall period the plane is recovered to the normal flight level. During pull up and recovery the test equipment has to stand an acceleration of about $2g_0$. In this way it is possible to perform 30 parabolas during one flight each providing an experimental time of about 20 s under microgravity conditions.

3. Possibilities for combined terrestrial and microgravity investigations on viscoelastic behaviour in ferrofluids

One of the actual research fields in the frame of investigations of magnetic fluids is the search for viscoelastic behaviour in these fluids. As a prominent indicator for viscoelasticity in a fluid the existence of normal stress differences is often used. For magnetic fluids it has been predicted [11], that normal stress differences should appear if chain formation of the magnetic particles is present in the fluid. The strength of the effect should increase with the length of the particle chains. Rheological investigations using a special dedicated rheometer for magnetic fluids [12] have shown an evidence for formation of chains even in magnetite based commercial magnetic fluids [13]. Thus it seems to be reasonable to search for normal stress differences in magnetic fluids under influence of magnetic fields, and to try to investigate the dependence of these deviations from normal newtonian behaviour as a function of the strength of the magnetic field. The data obtained that way is expected to give a good basis for the proof of different theoretical approaches in this field [11.14].

For the detection of normal stress differences we focus on a combined terrestrial and microgravity research program. With rheological measurement using the rheometer described in Ref. [12] one can determine the first normal stress coefficient. The small shear rate limits v_{i0} of the normal stress coefficients $v_i = 1/\dot{\gamma}^2 N_i$ are defined by

$$v_{i0} = \lim_{\dot{\gamma} \to 0} \frac{1}{\dot{\gamma}^2} N_i, \quad i = 1,2$$
(1)

with the normal stress differences N_{i} , and the shear rate $\dot{\gamma}$. For the determination of v_{10} the rheometer has to be operated in oscillating mode subjecting the fluid to a timedependent shear γ and registrating the resulting stress τ . Shear and stress will show a phase difference δ (see Fig. 1) which is 90° for a pure viscous fluid and 0° for a rigid body. For a viscoelastic liquid showing normal stress differences δ will take a value between 90° and 0°. For small oscillation frequencies ω , the phase difference can be used as a measure for v_{10} . More precisely the determination of the low frequency limit of the dynamic module $G'(\omega)$

$$\lim_{\omega \to 0} \frac{G'(\omega)}{\omega^2} = \lim_{\omega \to 0} \frac{1}{\omega^2} \frac{\tau_0}{\gamma_0} \cos \delta = \frac{1}{2} v_{10},$$
 (2)

where τ_0 and γ_0 are the amplitudes of stress and shear, respectively, allows to estimate the first normal stress coefficient.

For the second normal stress coefficient the determination is much more complicated, since the effect is expected to be too small for all conventional test methods like, e.g. direct force measurement. A possibility to obtain information on v_{20} is the investigation of the behavior of a free ferrofluid surface at a rotating axis immersed in the liquid (see Fig. 2).

In a normal Newtonian liquid the fluid surface will bend downward due to action of centrifugal forces. If the liquid exhibits normal stress differences, the centrifugal forces will be countered by a force radially inward which can drive the surface to bend upward like it is shown in Fig. 2. This so



Fig. 1. Definition of the important figures in oscillating rheological investigations for determination of v_{10} .



Fig. 2. Principle sketch of the behavior of a viscoelastic fluid at a rotating axis.

called Weissenberg effect – named after the austrian physicist Carl Weissenberg (*1893 † 1976) – is a measure for the combination $v_{10} + 4v_{20}$, since the height of rise of the fluid surface at the axis $h_{(r_0)}$ can be written in the form

$$h_{(r_0)} = \frac{r_0^2 \omega_0^2}{g} \left[-\frac{1}{2} + \frac{v_{10} + 4v_{20}}{\rho r_0^2} \right]$$
(3)

if effects of surface tension can be neglected. Here r_0 denotes the radius of the rotating axis, ω_0 its rotation frequency, ρ the density of the liquid and g the gravitational acceleration acting on the fluid. Thus, if it is possible to determine the height of the fluid surface, and if one obtains information on v_{10} from the rheological investigations described before, one can extract v_{20} from the data on Weissenberg-effect, obtaining detailed information on a certain class of viscoelastic properties of ferrofluids.

As mentioned before, normal stress differences are small in commercial magnetic fluids. Therefore it is impossible to observe the change of height of the fluid surface as a function of the strength of a magnetic field applied to such a ferrofluid under normal laboratory conditions. As one can see from Eq. (3) a reduction of gravitational acceleration in microgravity experiments should result in a remarkable amplification of the Weissenberg-effect since h scales proportional to 1/g. For parabolic flight experiments that would result in an amplification in the order of 100. As shown previously, surface tension effects can reduce the amplification effect [15]. But for extremely weak viscoelastic fluids, showing no rise of the fluid surface under terrestrial conditions, the 1/g-amplifictaion law could be verified [16], opening a possibility to observe even small effects of normal stress differences.

For magnetic fluids one expects, that the normal stress differences grow with increasing magnetic field strength. Therefore the height of the fluid surface at the rotating axis should increase with field strength too. As seen from (3) it should be negative for H = 0, since the fluid does not show viscoelastic behavior and in particular normal stress differences for vanishing magnetic field. With increasing field strength, the fluid surface should rise, get flat and finally at strong fields it should show a positive elevation. Of particular interest is the magnetic field strength H^* , for which the fluid surface is flat. For this surface elevation the expression in brackets in (3) is equal to zero, thus, for known radius of the rotating rod, it allows the direct determination of $v_{10} + 4v_{20}$. For magnetic fluids one has to observe the fact that this critical magnetic field strength H^* will depend on the shear rate and therefore on the rotation speed of the rod, since the length of the chains of magnetic particles will reduce with increasing shear rate [13,14].

4. Experimental investigations on Weissenbergeffect under reduced gravity conditions

To perform experiments like they have been discussed in principle above, we have build up a flight module for parabolic flight experiments using the ESA Airbus A 300. Fig. 3 shows the central part of flight hardware. It consists of a fluid cell, which is 3 bar pressure tight for safety reasons. The cell contains the rotating rod, driven by a motor coupled to the rod by magnetic coupling. This setup is surrounded by four magnetic field coils providing a magnetic field with a homogeneity appropriate to avoid side effects by small magnetic field gradients under microgravity conditions [17]. The behavior of the liquid is observed with a video camera.



Fig. 3. Sketch of the central part of the flight hardware for experimental investigation of Weissenberg-effect in parabolic flights.

The cell has been equipped with wetting barriers to prevent disturbations due to surface tension effects according to former experiences in experiments on Weissenberg-effect in parabolic flights [16]. The described setup is mounted in a flight rack containing power supplies, video recording system and electronic experiment control as well as seven additional fluid cells containing different fluids and rods of various diameters.

With this hardware we have carried out a series of flight experiments to prove the appearance of normal stress differences in a commercial magnetite based ferrofluid. First of all we have confirmed that surface tension effects do not influence the behavior of the free fluid surface under microgravity conditions by observing it with the axis at rest. Since no changes of the fluid surface have been observed one can state, that no wetting disturbations are present and therefore, that the observed behavior of the fluid is purely determined by centrifugal and normal stress forces. Fig. 4 shows the height of the fluid surface as a function of magnetic field strength in the ferrofluid APG 513A produced by ferrofluidics for loudspeaker applications. The fluid contains 7.2 vol% of magnetite particles with a mean diameter of about 10 nm. The rod with a radius of $r_0 = 5 \text{ mm}$ rotated with a frequency



Fig. 4. The height of rise of a free surface of APG513A at a rotating rod as a function of magnetic field strength.

 $f_0 = \omega_0/2\pi = 4$ Hz corresponding to a shear rate $\dot{\gamma} = 0.57 \text{ s}^{-1}$.

As it is clearly seen, the fluid surface rises with increasing magnetic field strength from strongly negative to slightly positive values crossing h = 0 mm at a critical field of about $H^* = 26.3$ kA/m. For the maximum available magnetic field strength the liquid shows a small Weissenberg-effect with a rise of the fluid surface at the rotating rod in the order of 0.25 mm. From the diameter of the rod one can obtain the value for the combination $v_{10} + 4v_{20} = 1.25 \times 10^{-5}$ kg/m.

Further experiments determining the first normal stress coefficient from rheological investigations are currently under preparation.

5. Discussions

It has been shown that microgravity experiments can provide unique experimental conditions allowing the investigation of magnetic effects in ferrofluids covered by gravitational action in normal terrestrial examinations.

For the particular problem of investigation of normal stress differences, it has been discussed that a combination of terrestrial and microgravity experiments can enable a deeper insight into the viscoelastic properties of magnetic fluids and their dependence on magnetic field strength. The results from the parabolic flight experiments have illustrated, that magnetic fields induce normal stress differences in ferrofluids. It was shown that Weissenberg-effect can be observed in stable ferrofluids containing nano-sized magnetite particles. The appearance of the effect, together with the predictions in Ref. [8] strengthens the assumption of chain formation of magnetic particles or primary agglomerates [11] under influence of magnetic fields, as it was already assumed on the basis of the results in Ref. [10].

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