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Hindrance of thermomagnetic convection by the magnetoviscous effect

H. Engler [*], A. Lange, D. Borin, S. Odenbach

Institute of Fluid Mechanics, Chair of Magnetofluiddynamics, George-Bähr-Str.3, 01062 Dresden, Germany

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ABSTRACT

The material and flow properties of magnetic fluids – so called ferrofluids – can be significantly influenced by magnetic fields. Two of the most prominent phenomena are the thermomagnetic convection and the magnetoviscous effect. To date, these magnetic field induced effects have always been studied separately, although they can affect each other under certain conditions. With the help of magnetic fields it is possible to induce a convective heat flow in ferrofluids. In earlier studies it is assumed for that case that the classical material properties are constant. However the magnetoviscous effect describes an increase in viscosity under magnetic influence, which occurs in ferrofluids with a certain contingent of large particles. In this paper, experimental results concerning the influence of the magnetoviscous effect on thermomagnetic convection are shown and discussed. Ferrofluids with and without significant magnetoviscous effect have been investigated.

1. Introduction

Thermomagnetic convection [1–5] is one of the most popular heat and mass transfer phenomena in magnetic fluids. The driving force behind this convection is a magnetic body force which arises in a horizontal layer of magnetic fluids if a homogeneous magnetic field is applied perpendicular to the fluid layer and additionally a temperature gradient over the fluid gap exists.

Stabilizing effects of thermal diffusivity and viscous friction act contrary to this destabilizing force. All three effects are combined in the magnetic Rayleigh number which is given by

$$Ra_m = \frac{\mu_0 \cdot K \cdot \nabla H_i \cdot \Delta T \cdot d^3}{\kappa \cdot \eta} = \frac{\mu_0 \cdot K^2 \cdot \Delta T^2 \cdot d^2}{\kappa \cdot \eta}, \tag{1}$$

where ∇H_i and ΔT denote the inner magnetic field gradient and temperature difference over the fluid gap with the thickness d, μ_0 is the vacuum permeability. K symbolizes the pyromagnetic coefficient, κ the thermal diffusivity, and η the dynamic viscosity of the fluid. It is assumed that κ and η are constant in contrast to K which depends on the magnetic field, see Fig. 1. If the magnetic Rayleigh number surpasses a certain critical value, a convective flow, solely driven by the magnetic body force, sets in. This phenomenon is called thermomagnetic convection. The established temperature difference over the gap by heating from below causes also a density gradient which produces an additional body force. This force is able to destabilize the fluid layer too. This well known heat and mass transfer phenomenon is established as thermal convection. As before the condition of the thermal convection can be characterized by the dimensionless thermal Rayleigh number which is given by

$$Ra = \frac{\beta_T \cdot g \cdot \rho \cdot \Delta T \cdot d^3}{\kappa \cdot \eta}, \tag{2}$$

where g is the acceleration of gravity, β_T the thermal expansion coefficient, and ρ the density of the fluid.

For a horizontal magnetic fluid layer which is subjected to a magnetic field and a temperature difference the Rayleigh numbers for thermomagnetic and thermal convection can be added to a total Rayleigh number $Ra_{TOT} = Ra_m + Ra$ [3] due to the co-directional orientation of both driving forces. The characteristic of heat transfer changes from a conductive to a convective state if Ra_{TOT} exceeds a critical value. In the case of pure thermal convection Ra_{crit} is 1708 [2] for a horizontal fluid layer bounded by rigid boundary plates with high heat conductivity. In this paper we assume that Ra_{TOT} is 1708 as well.

Fig. 2 shows in principle the heat flux through the horizontal fluid layer as a function of ΔT for different values of the applied magnetic field strength. The kink in the slope of the curve for H = 0 kA/m signalizes the transition point, where the characteristic of the heat flux changes from conduction to convection. This point determines the critical temperature difference ΔT_{crit}. Without an

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[*] Corresponding author.
E-mail addresses: harald.engler@tu-dresden.de, harald_engler@yahoo.de (H. Engler).

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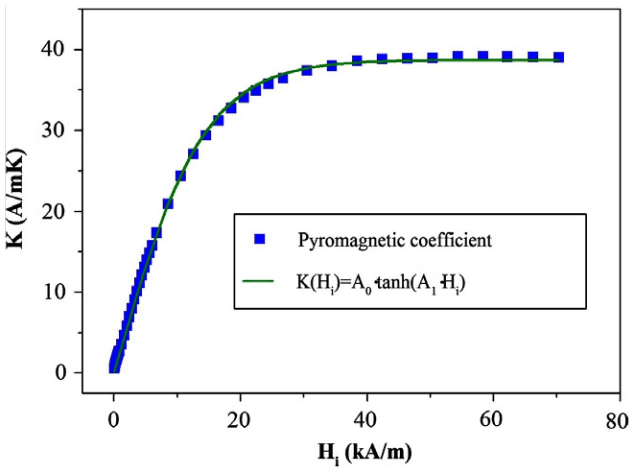

Fig. 1. This diagram shows the relation between the pyromagnetic coefficient and the inner magnetic field for the ferrofluid sample APG 513A_SMALL which is similar to Ferrofluid sample APG 513A_LARGE (not shown). The values of the variables of the fit are A_0 = 38.73 A/mK and A_1 = 0.07 m/kA.

applied magnetic field the heat flux is only driven thermally, so that ΔT_{crit} corresponds to $Ra_{TOT} = Ra = Ra_{crit}$. In the presence of a magnetic field the heat flux increases in general due to the additional heat flux induced by the magnetic body force. In this situation both mechanisms - thermomagnetic and thermal convection – are working simultaneously, so that ΔT_{crit} corresponds to $Ra_{TOT} = Ra + Ra_m = Ra_{crit}$.

In the case, where a convective flow is driven by the magnetic body force, it is assumed that the material properties of ferrofluids are constant and independent from the applied magnetic field with the obvious exception of the magnetization [5]. However, former theoretical investigations on ferrofluids have shown that the magnetic field can indeed influence the material properties undercertain conditions. The most popular effect in ferrofluids is the magnetoviscous effect [6], i.e. the change of the viscosity in the presence of a magnetic field. By applying a magnetic field to the ferrofluid, the direction of the magnetic moments of the particles aligns with the direction of the field. If the alignment reaches a certain value, the magnetic interparticle interaction of the particles are strong enough so that the particles are able to build chainlike structures which cause a strong increase of viscosity.

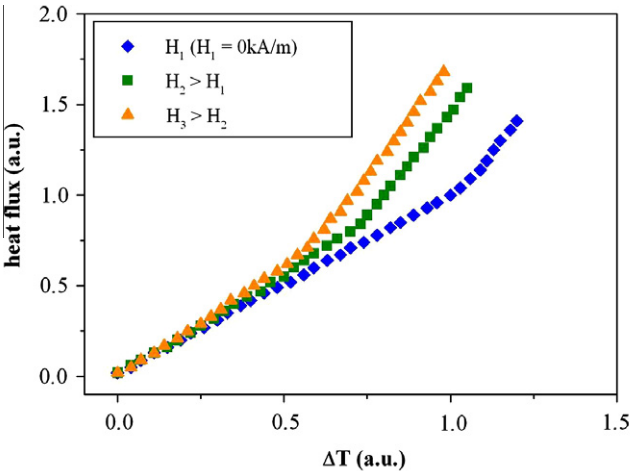

Fig. 2. The heat flux is shown as a function of the temperature difference. The kink in the slope indicates a characteristic change of the heat flux from conduction to convection. In general the heat flux increases with the increase of magnetic field strength.

For the above described mechanism it is assumed that only large particles are present which follow the Brownian relaxation process [14]. For this process the direction of the magnetic moments are fixed within the structure of the particles so that the whole particle has to rotate if it aligns with the magnetic field direction. The magnetoviscous effect is especially significant for high concentrated ferrofluids with a contingent of large particles [7–9]. For this situation a sufficient amount of large particles are present to build chainlike structures which have a major influence on the fluid properties and viscosity as well.

To determine the magnitude of the magnetoviscous effect an interaction parameter λ has to be introduced which is defined as the ratio between magnetic interaction energy between two particles which are in contact and their thermal energy [1]. To avoid the direct contact and therefore the van der Waals forces between two particles they are coated by a surfactant layer. These additional geometric boundary conditions are considered by introducing a modified interaction parameter [10]

$$\lambda^* = \lambda \cdot \left(\frac{d_{particle}}{d_{particle} + 2 \cdot s}\right)^3 = \left(\frac{\mu_0 \cdot M_0^2 \cdot V}{24 \cdot k_B \cdot T}\right) \cdot \left(\frac{d_{particle}}{d_{particle} + 2 \cdot s}\right)^3, \qquad (3)$$

where M_0 denotes the spontaneous magnetization of the magnetic material, V is the volume of the magnetic particle, k_B the Boltzmann constant and T the absolute temperature. Furthermore $d_{particle}$ denotes the mean diameter of the particles and s is the thickness of the layer of the surfactant. If the value of λ^* is significantly larger than 1 the interparticle interactions are strong enough to build chainlike structures, i.e. the energy of the interparticle interaction is higher than the thermal energy.

Here, we investigate how thermomagnetic convection is affected by the magnetoviscous effect. Both phenomena – thermomagnetic convection and magnetoviscous effect – are well known but former experimental and theoretical investigations have used that both effects are independent from each other. A usual assumption is that the fluid properties are independent from the applied magnetic field, although it has been proven that in certain circumstances the magnetoviscous effect causes an amplification of η if a magnetic field is applied. An increase of η strengthens the stabilizing viscous force, see Eq. (2), which is why the magnetoviscous effect hinders the onset of thermomagnetic convection. Therefore it is expected that for ferrofluids with high values of λ^* the critical temperature difference is shifted to higher values.

2. Experimental setup

The magnetic field which is necessary to investigate the field influence on heat and mass transfer phenomena in ferrofluids is generated by two pairs of Helmholtz coils. The coils are placed in a Fanselau arrangement which ensures a high homogeneity of the field. The maximum magnetic field strength which can be generated by the coils is 25 kA/m.

The actual measuring cell is located in the centre of the Fanselau arrangement and permits to measure the heat flux through the horizontal fluid layer as well as the temperature difference ΔT over the fluid gap. The fluid gap has a height of 4 mm and a diameter of 88 mm which leads to an aspect ratio of 22. For such high aspect ratio the influence of the vertical boundaries on the heat flux phenomena in the layer is insignificant [11]. The material of the horizontal boundary plates which cover the fluid gap on the top and on the bottom is copper, with a heat conductivity of 378 $\mathrm{W\,K^{-1}\,m^{-1}}$. The temperature in the copper plates is measured by thermistors from the company BetaTherm Sensors, Germany. They have a response time of 30 ms and a basic resistance of 20 kΩ at a temperature of 20 °C. The sensitivity of the thermistors is 2000 Ω/K. That means if the resistance of the thermistors can be measured with an accuracy of 1 Ω, which is a typical resolution for laboratory measurements, the temperature resolution of the thermistors is in the order of 10^{-3} K. The thermistors are coated with a glass cylinder which has a length of 1.38 mm and a diameter of 0.38 mm. The cables for the data transmission are isolated. The maximum temperature difference ΔT which can be applied to the fluid gap is $\Delta T \approx 35$ K. A more detailed description of this part of the experimental set up is given in [4].

To measure the heat flux through the horizontal fluid layer the set up is equipped with a specialized heat flux sensor (provided by the company Ahlborn, Germany). The sensor consists of a chain of thermal elements which are embedded in a resin of polyester in a meander-like structure. The active area of the circular sensor has a diameter of 23 mm, whereas the total diameter is 33 mm. The outer region without an active measuring ensures that side effects do not influence the measurement of the heat flux in the internal area. The height of the sensor is 1.5 mm. The sensor is located between two copper plates in the lower part of the cell. One of the plates is in direct contact with the ferrofluid layer. The heat flux density is 114 W/mm^2 if a thermo-voltage of 1 mV is displayed on the heat flux sensor. The measuring region ranges from 0.5 W/m^2 to 3000 W/m^2.

The boundary layers are coated with a heat-conductive paste to improve the contact between the sensor and the copper plates. In place of the heat flux sensor a thermal resistance layer is located between two copper plates in the upper part of the measuring cell. This layer has a similar heat conductivity as the heat flux sensor in order to keep the symmetry of the cell.

For the investigation of the influence of the applied magnetic field on the viscosity, a shear-controlled rheometer described in [13] was used with some technical modifications. The magnetic field is generated by a pair of Helmholtz coils with a maximum field strength up to 40 kA/m. The field homogeneity in the center of the coils is better than 95%. The measurement cell of the rheometer consists of a cone-plate geometry and allows shear rate variations in the range from 10^{-4} s^{-1} to 10^{3} s^{-1}.

The ferrofluids which are used for the investigation are commercial ferrofluids called APG 513A (Ferrotec). The magnetic particles of the fluids are made of magnetite (6.3 vol.%) and the carrier liquid is a synthetic ester. The saturation magnetization M_S is measured to 28.2 kA/m at room temperature of 20 °C and the pyromagnetic coefficient K is 35.3 A m^{-1} K^{-1} for a field strength of 25 kA/m. Further basic fluid properties are the thermal expansion coefficient $\beta_T = 6.4 \cdot 10^{-4}$ 1/K, the thermal diffusivity $\kappa = 1.1 \cdot 10^{-7}$ m^2/s, the density $\rho = 1345$ kg/m^3, and the viscosity $\eta_{(T=20\ ^\circ\mathrm{C})} = 195$ mPa s.

Two different samples of the ferrofluid APG 513A are used for the investigations, which have in principle the same basic and magnetic properties as shown above. The exception is the size distribution of the particles which is important for the formation of the magnetoviscous effect. The fluid sample APG 513A_LARGE has a wide particle size distribution. Thus a significant number of magnetic particles with a diameter of 14 nm or larger is present in this sample. The second sample APG 513A_SMALL contains particles, where most of them have a diameter close to the mean value of 10 nm.

3. Experimental results

In the first step of the experimental investigations we have determined the influence of an applied magnetic field on η of the fluid for both samples. In Fig. 3 the viscosity is shown as a function of the applied magnetic field strength for a shear rate of $\dot{\gamma} = 0.1$ s^{-1}. For the sample APG 513A_SMALL only a slight increase of η with increasing field strength can be observed. For this situation the ratio of $\Delta\eta$ to η_0 is about 0.5 for a field strength of 30 kA/m. Due to the size distribution of the particles the magnetic interparticle interaction parameter λ^* is smaller than 1 and the magnetoviscous effect can hardly influence η.

The dependence of η on a magnetic field changes dramatically for the ferrofluid sample APG 513A_LARGE. The ratio of $\Delta\eta$ to η_0 is about 4.4 for a field strength of 30 kA/m. This sample has a high concentration of particles with a diameter larger than the mean diameter of 10 nm so that the magnetic interparticle interaction is strong enough to build chainlike structures. Actually a small amount of the particles in this sample with a volume concentration of $\Phi = 0.8\%$ has a mean diameter of 16 nm, leading to a magnetic interparticle interaction parameter of $\lambda^* = 2.87$ [13].

In the second step we have measured the progress of the heat flux through a horizontal ferrofluid layer heated from below as a function of the applied ΔT over the fluid gap. We also measured the heat flux for different strengths of the applied magnetic field. To keep a mean temperature in the gap of 40 °C and therefore to keep the fluid properties which are sensitive to the temperature constant, ΔT was increased stepwise by a symmetrical temperature change of 1 K on the bottom and on the top of the measuring

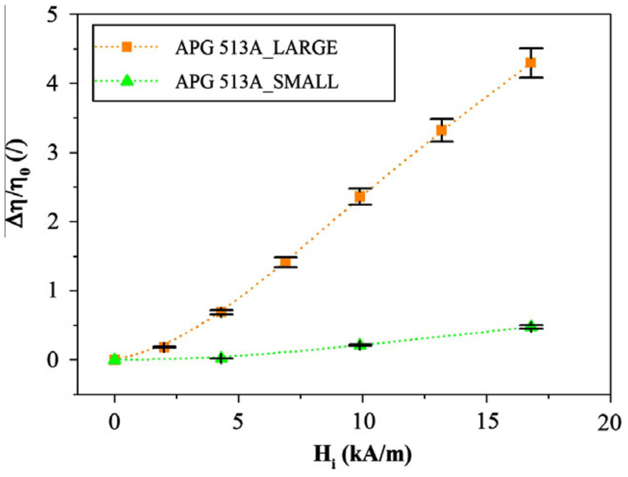

Fig. 3. The shape of the fit-equation is $\Delta\eta/\eta_0 = P_1 \cdot H_i^{P_2}$. Thereby the values of the variables of the fits are $P_1 = 0.13$ and $P_2 = 1.24$ for the sample APG 513A_LARGE and $P_1 = 0.003$ and $P_2 = 1.8$ for sample APG 513A_SMALL.

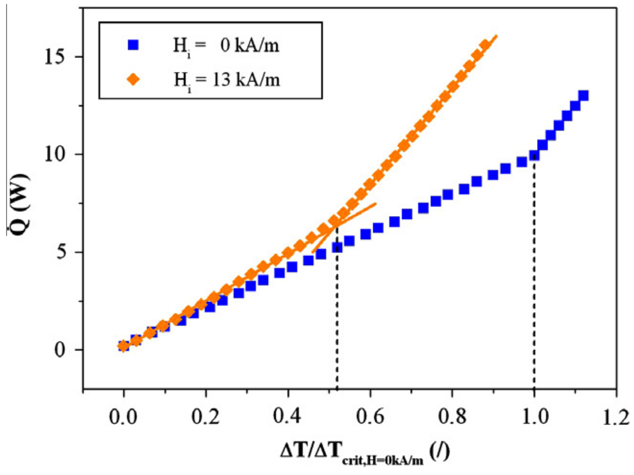

Fig. 4. Heat flux as a function of the temperature difference over the fluid gap for the ferrofluid sample APG 513A_SMALL. The change in the slope denotes the transition from conduction to convection.

cell. The heat flux changes from a conductive to a convective state if ΔT_{crit} is passed. At this ΔT_{crit} the slope of the heat flux curve changes significantly, which can be seen in Fig. 4 for the sample APG 513A_SMALL. Without an applied magnetic field, where only the buoyancy forces drive the convection, ΔT_{crit} is 32 °C, which is normalized to 1. If we apply the maximum field strength of 25 kA/m – equivalent to H_i = 13 kA/m – the heat flux in the fluid layer increases generally due to the fact that the induced magnetic force supports the heat flux additionally. Furthermore in Fig. 4 is shown that for this combined heat flux the normalized critical temperature difference has a value of about 0.5 that means it is decreased by a factor of nearly 2.

Fig. 5 shows ΔT_{crit} as a function of the applied magnetic field strength, where the values of ΔT_{crit} for H = 0 kA/m and H = 25 kA/m correspond to Fig. 4. Additionally Fig. 5 shows ΔT_{crit} for further values of the magnetic field strength which are not shown in Fig. 4. In Fig. 5 is also presented the analytical determination of ΔT_{crit} (line) which agrees well with the experimental data by using the parameters of the fluid and of the experimental setup. For the analytical determination Eqs. (1) and (2) are used whereas the assumption is employed that the value for $Ra_{TOT} = Ra_m + Ra = Ra_{crit}$ is 1708 [5]. This value for Ra_{crit} is kept constant for this calculation, so that

$$\Delta T_{crit} = f[K(H), Ra_{TOT} = 1708]. \qquad (4)$$

Actually $Ra_{crit} = Ra_m + Ra$ can be transformed in a quadratic equation for ΔT_{crit}. The solution is given by

$$\Delta T_{crit} = -\frac{d\cdot\beta_T\cdot g\cdot\rho}{2\cdot\mu_0\cdot(K_{H_i})^2} + \sqrt{\frac{d^2\cdot\beta_T^2\cdot g^2\cdot\rho^2}{4\cdot\mu_0^2\cdot(K_{H_i})^4} + \frac{1708\cdot\kappa\cdot\eta}{d^2\cdot\mu_0\cdot(K_{H_i})^2}}. \qquad (5)$$

For the calculation with Eq. (5) the values of the fit presented in Fig. 1 are used for $K(H)$.

With the help of Eq. (5) it can be seen that by increasing of the magnetic field strength, and therefore an increasing of the heat flux driven by the magnetic force, ΔT_{crit} has to decrease. The analytical results of Eq. (5) are also shown in Fig. 5 as the red line. The experimental results from the measurements with APG 513A_SMALL confirm the expected behaviour and the results in [3].

As mentioned before, APG 513A_LARGE presents a sample with a wide particle size distribution and the concentration of large particles are high enough to lead to a significant magnetoviscous effect [6]. Without an applied magnetic field the change of heat flux with ΔT shows the same behaviour as the ferrofluid sample APG 513A_SMALL, i.e. the heat flux increases with an increase of ΔT. In Fig. 6 the characteristic kink of the slope in the heat flux curve can also be seen and gives the value of ΔT_{crit} which is again 32 °C. The situation changes dramatically if a magnetic field with a strength of 25 kA/m is applied. For this sample the magnetic field causes an additional heat flux as well as an increase of η. Corresponding to the definition of Ra_m, see Eq. (1), the numerator in this equation represents mainly the magnetic force leading normally to an increase of Ra_{TOT}. Since η is part of the denominator, both effects counteract, leading effectively to a decrease of the heat flux. Thus higher values of ΔT_{crit} are needed to reach Ra_{crit}. Fig. 7 shows the measured values of ΔT_{crit} as a function of the strength of the applied magnetic field. It can be seen that ΔT_{crit} for $H \neq 0$ kA/m is higher than for H = 0 kA/m. Remarkably, it seems that the values of ΔT_{crit} are almost independent of the strength of the applied magnetic field and they are nearly constant. This behaviour is totally different to the behaviour of the sample APG 513_SMALL and it is not yet understood.

We have already seen that APG 513A_LARGE shows a significant magnetoviscous effect, see Fig. 3, so that η increases remarkably

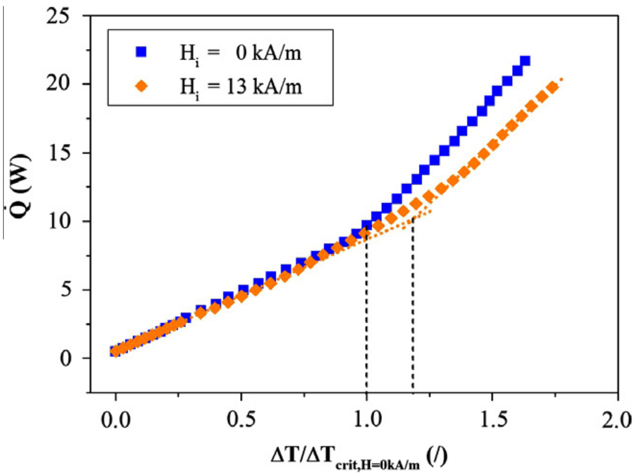

Fig. 6. Heat flux as a function of the temperature difference for the ferrofluid sample APG 513A_LARGE. The sample contains a contingent of large particles leading to structure formations by applying a magnetic field and an increase of the viscosity.

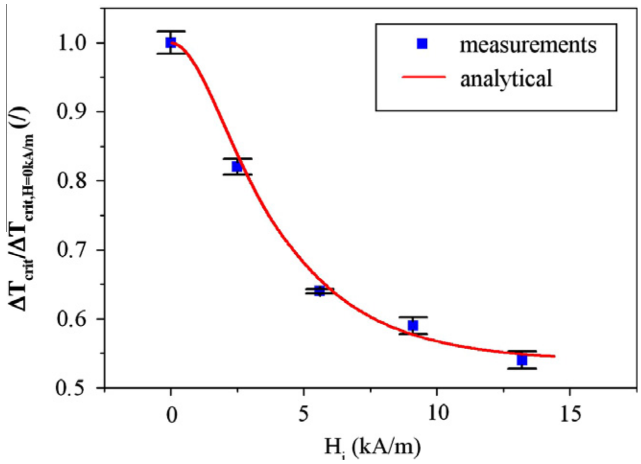

Fig. 5. ΔT_{crit} as a function of the applied field strength for the ferrofluid sample APG 513A_SMALL. Furthermore, analytical data points are presented using the parameters of the material and the geometry of the experimental setup.

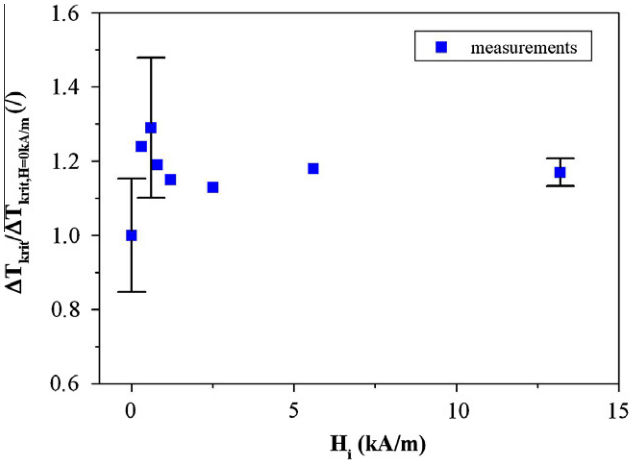

Fig. 7. Behaviour of ΔT_{crit} depending on the applied magnetic field strength of the sample APG 513A_LARGE. ΔT_{crit} is almost constant and independent of the field strength.

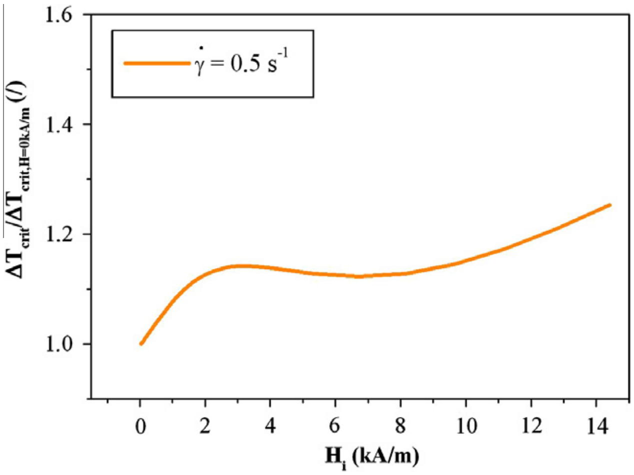

Fig. 8. Calculation of the critical temperature difference ΔT_{crit} for a shear rate of $\dot{\gamma} = 0.5\ s^{-1}$. In this case the increase of η depending on the magnetic field strength is smaller because of the higher shear rate. It breaks the chain-like structures formed by the particles in contrast to the lower shear rate. This effect is called shear thinning [6].

with magnetic field strength. For the analytical determination of ΔT_{crit} in Fig. 8 a modified Eq. (5) is used. The assumption of $Ra_{TOT} = 1708$ leads to a good agreement for the ferrofluid sample APG 513A_SMALL. So we assume that for APG 513A_LARGE $Ra_{TOT} = Ra_m + Ra = Ra_{crit}$ is 1708 as well and kept constant for the determination. Finally ΔT_{crit} is given by

$$\Delta T_{crit} = f[\eta(H), K(H), Ra_{TOT} = 1708]. \tag{6}$$

In comparison to Eqs. (4) and (6) has a magnetic dependence of the pyromagnetic coefficient K and additionally of the viscosity η. This additionally situation is introduced in the modified Eq. (5). As mentioned before the magnetic field causes an increase of the magnetic force, especially the pyromagnetic coefficient K leading to a decrease of ΔT_{crit}. In the case of APG 513A_LARGE the magnetic field causes also an increase of the viscosity which has a stabilizing character, so that ΔT_{crit} is shifted to higher values. Both phenomena, the pyromagnetic coefficient K and the viscosity, counteract each other, which leads to an almost entire compensation. This leads partly to a nearly constant value of ΔT_{crit} in Fig. 8 which is comparable with the measurements in Fig. 7. For the calculation of the curve of ΔT_{crit} in Fig. 8 the value of η for a shear rate of $\dot{\gamma} = 0.5 s^{-1}$ is used.

In order to estimate the shear rate occurring in the flow field of convective heat transport it is assumed that a simple geometry of the convection rolls as shown in Fig. 9 is present. Additionally, it is supposed that the gradient of velocity inside the rolls is linear. With these assumptions a constant shear rate exists across the plane shear flow. The shear rate is then given by $\dot{\gamma} = \partial v/\partial y \approx \Delta v/\Delta y$. For a fully developed convection flow in a horizontal layer of fluid, velocities of about 1 mm/s can be expected [14]. Locating this velocity close to the boundaries of the layer which has a thickness of 4 mm results in a shear rate of 0.5 s^{-1}.

4. Conclusion

In this paper the influence of the magnetoviscous effect on the behaviour of the onset of thermomagnetic convection has been investigated. As set up for this investigation a horizontal layer of magnetic fluid subjected to a homogeneous magnetic field and a temperature gradient, both oriented vertically, is used. The investigations presented here show that for a ferrofluid with a high concentration of large particles the critical temperature difference, when the characteristic of the heat flux changes from conduction to convection, is shifted to higher values in the presence of a magnetic field.

For the measurements of the magnetoviscous effect we used a shear controlled rheometer with a cone-plate geometry of the measuring cell. The measurements have been done amongst others with a shear rate of 0.1 s^{-1} and field strength of up to 25 kA/m which correspond to an inner magnetic field strength of 13 kA/m. For the maximum field strength we have an increase of η by a factor of 4.4 for the ferrofluid sample APG 513A_LARGE, whereas η increases only slightly in APG 513A_SMALL. The influence on the thermomagnetic convection has been determined with a measuring cell which is able to measure the heat flux as well as the temperature difference over the fluid gap.

The experiments for the thermomagnetic convection have shown that for the ferrofluid sample APG 513A_SMALL without an applied magnetic field the critical temperature difference is 32 °C and decreases by a factor of 2 if the maximum field strength of 25 kA/m is applied. In this situation the induced magnetic body force causes an additional heat flux which leads to a decrease of ΔT_{crit}. The experimental results are well confirmed by an analytical calculation of ΔT_{crit}.

The behaviour of ΔT_{crit} changes if we use the ferrofluid sample APG 513A_LARGE. This sample contains a comparably high volume fraction of large particles which are able to form chain-like structures [13]. This structure formation leads to an increase of η in the presence of a magnetic field. Without an applied magnetic field ΔT_{crit} is 32 °C, which agrees with the first ferrofluid sample. In the case of $H \neq 0$ kA/m the values of ΔT_{crit} increase to a value which is higher than ΔT_{crit} = 32 °C and it is nearly constant for any applied magnetic field strength. To get a first understanding of this behaviour we have considered the equation of Ra_m. The pyromagnetic coefficient $K(H)$ which is mainly responsible for the magnetic force destabilizes the thermomagnetic convection leading to a decrease of ΔT_{crit} with increasing H. Simultaneously the magnetoviscous effect induces an increase of the viscosity which has a stabilizing effect on the thermomagnetic convection. This entails an increase of ΔT_{crit}. Effectively, the pyromagnetic coefficient $K(H)$ and the increase of viscosity $\eta(H)$ counteract each other, which leads partly to a compensation. The dependence of ΔT_{crit} on H_i in Fig. 8 is calculated for a shear rate of 0.5 s^{-1} and shows large regions of constant values of ΔT_{crit} which gives a qualitative explanation of the measurements in Fig. 7.

Finally the question about the jump in the experimental data of ΔT_{crit} in Fig. 7 by applying a magnetic field is still open. A similar behaviour is observed for a Bingham fluid [15] characterized by a

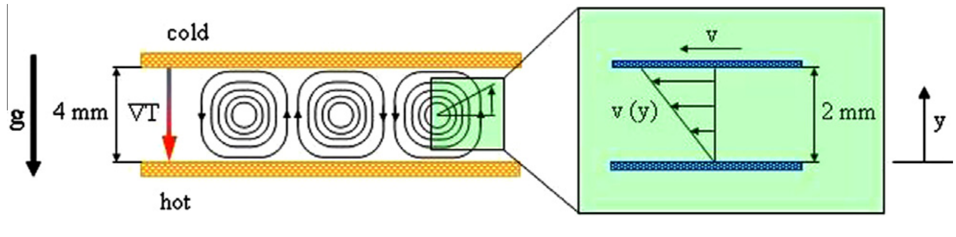

Fig. 9. Left: Sketch of the simple geometry of the convection rolls. Right: Illustration of the plane shear flow with a linear gradient of the velocity inside the rolls.

yield stress. The authors show that the onset of convection depends on the yield stress and leads to a change of Ra_{crit} and therefore to a change of the critical temperature difference. Experimental studies [12] performed recently have shown that the ferrofluid sample called APG_LARGE shows a yield stress in the presence of a magnetic field. It is necessary to carry out further studies to investigate the influence of a yield stress on the onset of thermomagnetic convection in more detail and to clarify whether this effect is capable to explain the sudden increase of ΔT_{crit} when switching on the magnetic field.

Acknowledgments

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