

Preface

Within the scope of this work we've investigated the magnetoviscous effects – i.e. the changes of viscous properties due to the action of magnetic fields – in so-called ferrofluids. These fluids, suspensions of magnetic nanoparticles in appropriate carrier liquids, show a pronounced increase of viscosity in the presence of moderate magnetic fields with strengths of the order of several tens of mT. Classically this effect is explained by the hindrance of the free rotation of magnetic particles – with a magnetic moment spatially fixed in the particle – in a shear flow due to magnetic torques trying to align the particles' magnetic moments with the magnetic field direction.

Starting from the classical theory by Mark Shliomis (Shliomis, 1972) we've performed a couple of experiments to validate the predictions of the theory. The use of relatively concentrated commercial magnetic fluids lead to the conclusion that the mentioned theory – developed for highly diluted fluids – is not able to give a quantitative description of the behavior of commercial fluids. The discrepancies have been attributed to the appearance of interparticle interactions between the magnetic particles.

Since the microscopic make-up of commercial ferrofluids is relatively complicated, and in particular parameters like the size distribution of the magnetic particles are not known precisely, a theoretical description of the microscopic reasons for the fluids' macroscopic behavior is impossible without further information. Therefore we've started a series of investigations shedding light on the viscous behavior of magnetic fluids in the presence of magnetic fields, stepwise reducing the number of relevant microscopic parameters to prepare a basis for sufficient modeling of concentrated ferrofluids.

As a first step in this development a specialized rheometer for the investigation of magnetic fluids has been designed. With this rheometer, allowing well-defined application of a magnetic field to a rheometric flow of ferrofluids, we've investigated the shear dependence of the magnetoviscous effect in commercial ferrofluids. These investigations showed that the field-dependent increase of viscosity reduces with increasing shear rate. On the basis of this result we developed a model, assuming that the formation of chains of magnetic particles dominates the magnetoviscous properties of magnetic fluids. The chains themselves represent large magnetic structures which lead to pronounced changes of viscosity if a field is applied. Furthermore, the rupture of the chains in a shear flow and the resulting reduction of the size of the magnetic structures is a starting point for the explanation of the observed shear thinning.

Since chains of magnetic particles can only be formed by particles exhibiting a sufficient interparticle interaction, and since this interaction depends furthermore

on the size of the particles, the next step had to be a clarification, whether the relatively small fraction of large particles in the suspension used is of major importance for magnetoviscosity in ferrofluids. These large particles exhibit – in contrast to the majority of particles with diameters of about 10 nm – sufficiently strong interaction to explain at least the appearance of chain formation.

To get an insight into these questions, we've performed experiments using ferrofluids with variable contents of large particles. In these experiments it was clearly shown that the magnetoviscous effect rises with an increasing amount of large particles. This leads to further input for the theoretical modeling. In an extended approach the ferrofluid is assumed to be a bidisperse system containing a large fraction of small particles, which do not directly contribute to magnetoviscosity, and a small fraction of large particles which form chains determining the field-dependent changes of viscosity. On the basis of these assumptions the magnetoviscous properties could be fitted quantitatively to the experimental data using methods of statistical physics. Thus, a first quantitative description of the microscopic reasons for the rheological behavior of ferrofluids was found, taking into account the effects to the formation of magnetic particle chains. The conclusion that chains exist in the fluids gives rise to the assumption that these fluids should exhibit viscoelastic effects too. To prove this, we finally carried out experiments on the Weissenberg effect, i.e. the climb of a free surface of magnetic fluids at a rotating axis, showing the field-dependent existence of normal stress differences in ferrofluids. Again, the experimentally found behavior could be explained by the formation and rupture of chains of magnetic particles in the fluid.

Thus – within the scope of this work – we've been able to develop a microscopic model of ferrofluids allowing a quantitative description of their rheological behavior, and to prove this model with numerous experimental results on field-dependent effects in ferrofluids rheology. On the basis of these results, information for the optimization of ferrofluids with respect to their magnetoviscous behavior can be obtained, leading to the synthesis of new ferrofluids. Such fluids with enhanced magnetoviscous properties may be used in the future development of devices using the magnetically induced control of viscous properties as an active part in technical applications like dampers or clutches.

Investigations like those described in this work require not only a certain time span to be performed but also the help and cooperation of numerous colleagues and the financial support enabling the research activities.

Thus I'd like to take the opportunity to express my gratitude to those helping me to do this research during recent years.

First of all I've to thank Prof. Dr.-Ing. H. J. Rath and Prof. Dr. K. Stierstadt for providing me with a working environment in Bremen as well as in former times in Munich that gave me the possibility of developing ideas and building up a research team able to explore this new and interesting field. Without these boundary conditions this wouldn't have been possible.

Furthermore my gratitude goes to my co-workers who were prepared to work even in difficult ways towards new scientific and technical goals: Dipl. Phys. H. Gilly for lively discussions during the time in Munich, Dipl. Phys. H. Störk who

built the first version of the ferrofluid rheometer in Wuppertal, and last but not least the members of the ZARM-ferrofluid team who participated in various experiments which led to the results presented, Dipl.-Ing. J. Fleischer, Dipl.-Ing. M. Heyen, Dipl.-Ing. K. Melzner, Dipl.-Ing. T. Rylewicz, Dipl.-Ing. S. Thurm and Dipl.-Ing. T. Völker.

Besides this I'm grateful to numerous colleagues and friends for fruitful and enlightening discussions. In this case it's nearly impossible to name all those who have been with me during the years, but I'd like to mention particularly: Prof. E. Blums, Prof. A. Zubarev and Prof. L. Vekas who were our guests in Bremen numerous times in the course of fruitful cooperations; Dr. K. Raj who provided us with the fluid series for the experiments concerning the influence of large particles; Prof. K. Stierstadt, Dr. H. W. Müller and Dipl.-Ing. Ch. Eigenbrod who helped me with deep and inspiring discussions; and numerous members of the German ferrofluid community who are helping to form a powerful research community on magnetic fluids.

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Since most of the work presented has an experimental character, the technical support provided by the workshop at ZARM and the Fallturm Betriebsgesellschaft was often of great importance to the success of our research. I'm especially grateful for this, since we often had to set extremely tight deadlines which were always observed.

Besides all the research work, these pages had finally to be written, and in this context I'd like to express my thanks to E. Renschen and C. Wieske for a lot of typing.

In general, the development of scientific activities is a part of life that can not be successful if it is not supported by an appropriate private environment. Many of the colleagues mentioned above have become real friends during the years, supporting me even in difficult times.

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