Study of structural changes in magneto-rheological fluids by magnetic field

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Abstract — The changes in structural arrangement in silicon oil based magneto-rheological fluids with carbonyl iron particles upon the effect of an external magnetic were studied by acoustic spectroscopy. The attenuation of acoustic waves was measured for a jump change of the magnetic field at temperature 20 °C for various magnetic particles concentrations. The relaxation effect for the acoustic attenuation after switching off the magnetic field and its decrease to the same value as for clean synthetic oil were observed. The change in attenuation coefficients of acoustic waves in magneto-rheological fluid versus angle between wave vector and direction of the applied magnetic field has been measured. From the measured anisotropy of attenuation coefficient structural change of magneto-rheological fluid in the magnetic field is evident. The observed influences of the magnetic field and concentration on the investigated liquids structure are discussed.

***Keywords** - acoustic spectroscopy; magneto-rheological fluid; acoustic attenuation; anisotropy.*

I. INTRODUCTION

Magneto-rheological (MR) systems are suspensions of magnetic micro-particles in a carrier fluid, usually mineral or silicon oil [Dong 12, Baranwall 12, Sedlacik 12]. In the absence of an external magnetic field, MR fluids are reasonably well approximated as Newtonian liquids. Their rheological properties (e.g. viscosity) are rapidly varied by applying a magnetic field. Under influence of the magnetic field there are interparticle interaction of suspended magnetic particles and their interaction with the magnetic field to form structures that have big influence on their shear deformation and flow. Their ability of changing shear stress in the presence of magnetic field gives rise to their many possible applications. In most engineering applications a simple Bingham plastic model is effective for describing the field-dependent fluid characteristics. So they can be applied in various fields of civil engineering, safety engineering, transportation and life science, in the design of brakes, dampers, clutches and shock absorbers systems [Dong 12, Kciuk 06, Baranwall 12].

The corrosion, oxidation, and abrasive properties of iron and iron alloys frequently used as optimal magnetic agents in MR fluids are, however, obstacles for their wider commercial usage. The modification of particle surface by chemical process has proven to be efficient to overcome this problem [Sedlacik 12, Dong 2012]. The improvement of properties of MR fluids by plasma-enhanced chemical vapour deposition of octafluorocyclobutane onto carbonyl iron (CI) particles using rotary plasma reactor was used in work [Sedlacik 12]. The results revealed successful fluorination of CI particles with a maximum of fluorine content of 2.9 %. The fluoropolymer film fabricated onto particles improved their corrosion protection and friction properties by 40%. The traditional CI particles can be replaced by amorphous alloy particles as the dispersed phase to prepare amorphous based MR fluid [Dong 12]. The amorphous particles have larger magnetization intensity and permeability at lower field levels as well as lower density. The MR fluid based on amorphous particles has better MR performance and sedimentation stability than that based on CI particles. Magneto-rheological fluids exhibit some advantages over typical electrorheological (ER) materials. In contrast to ER materials, MR fluids are more useful because the change in their rheological properties is more pronounced compared to ER fluids, i.e. an increase of yield stress is 20-55 times higher [Baranwall 12]. There are also electro-magneto-rheological fluids, which consist of particles responsive to both electric and magnetic field. The results from the molecular dynamic simulation studies have shown that under compatible electric and magnetic fields various types of structural formation and transition between them can exist [Wang 01]. The layers combine together to form thicker sheet-like structures, which finally relax into three-dimensional close-packed structures. The transitions between individual structures were observed when the electric field was applied as first following by the application of the magnetic field.

Currently, there are only few papers involved in the acoustic properties of MR fluids studied in this work. One of them is work by Baev [Baev 2015], who made similar acoustic measurement for higher concentration of magnetic particles, but with different type of MR fluid. Data analysis confirmed dependence of sound velocity and acoustic attenuation on the magnitude and direction of magnetic field in MR fluid. Their measurement also showed that interparticle interactions also depend on the processes in the interface boundary.

Similar properties and behavior under the external magnetic field as for MR fluids are observed for ferrofluids (FF) with magnetic nanoparticles [Kudelcik, Jozefcak, Skumiel]. The diameter and magnetic moments of magnetic particles in stable colloidal solutions are much lower than in MR fluids. Under the effect of an external magnetic field, the nanoparticles within FF become arranged into clusters, forming chains stiffening the liquid structure. The propagation of the acoustic wave in FF and change of acoustic attenuation by magnetic field was studied by several authors both theoretically and experimentally [Kudelcik, Sokolov].

II. EXPERIMENTAL

Carbonyl iron particles (HQ grade, BASF, Germany) were selected as magnetic agents for the investigation by acoustic methods. The main material characteristics of CI particles with HQ grade according to the manufacturer are following: spherical shape of particles with the average size of about 1 µm, non-modified surface, and content of α-iron >97%. The MR suspensions with the particle concentration of 1 and 2 wt.% in silicone oil (Lukosiol M50, viscosity $\eta_c = 50$ mPa·s, density $\rho_c = 0.970$ g·cm^{-3}, Chemical Works Kolín, Czech Republic) were prepared. Morphology and dimensions of the particles were observed using scanning electron microscope (SEM; Vega II LMU, Tescan, Czech Republic) operated at 10 kV. The magnetic properties of the samples were measured at room temperature using a vibrating sample magnetometer (VSM; Lakeshore, USA). Figure 1 shows the hysteresis cycles of CI particles under investigation. A soft magnetic behaviour of particles is presented since their increasing and decreasing branches of the hysteresis cycles are hardly distinguishable. The magnetization saturation in the field of 777 kA·m^{-1} is 174 emu·g^{-1}.

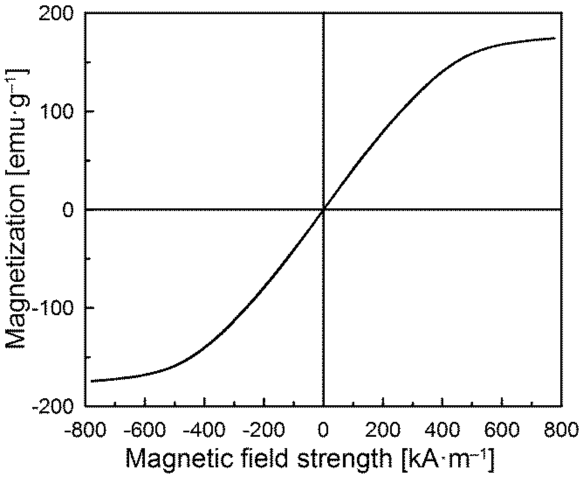

Fig. 1 Magnetization curve of CI particles under investigation.

The block diagram of the experimental equipment is shown in Fig. 2. The hf signal of frequency 4,9 MHz with pulse length 0,6 µs were generated by a pulse method using the MATEC Pulse Modulator and Attenuation Recorder, Model 7700. This signal was fed to piezoelectric transducer $LiNbO_3$ which generated the acoustic wave propagated through the MR fluid placed in the thermostatted closed measuring cell (1.5 x 0.9 x 1 cm^3, the temperature stabilized with an accuracy ±0.2 °C by JULABO Refrigerated & Heating Circulators F25-HE) inserted in an electromagnet underwent a multiple reflection between transducers. The echoes representing different paths after reflection and reaching a receiving transducer were received by the MATEC. The processed signals from the MATEC were displayed by the oscilloscope and from the first and the second echoes the acoustic attenuation in given magnetic fluid was evaluated and recorded by a computer. Computer also controlled current source for electromagnet.

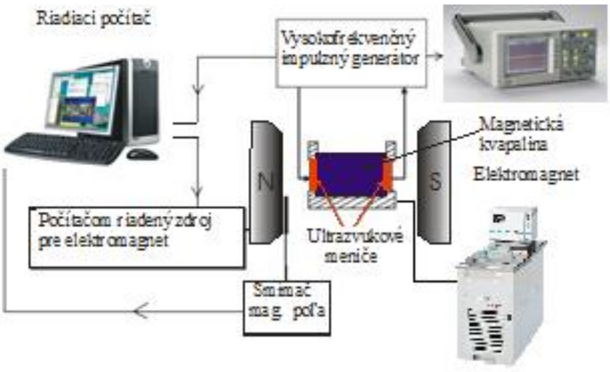

Fig. 2 Experimental setup.

III. RESULTS

Figure 3 present the changes of acoustic attenuation for a jump change of the magnetic field to 100, 200 and 300 mT for 1 wt.% MR fluid at 20 °C. The given value of magnetic field was kept constant during next 30 min. As can be seen from the measured development, the change of acoustic attenuation is immediate with the step change of the magnetic field. The value and development of acoustic attenuation depend on the magnitude of applied magnetic field. The most stable development was observed at 200 mT. For other magnitudes of magnetic field the acoustic attenuation wasn't stable, and decreased in time. At 100 mT the acoustic attenuation decreased under the value measured without the magnetic field. No significant effect on the value of acoustic attenuation, the relaxation process, was observed in the absence of the magnetic field. During next 20 min the value of acoustic attenuation didn't change except no stable development observed at 300 mT. After this time the acoustic attenuation exponentially decreased with time constant of about 500 s. The value of acoustic attenuation decreased to value as for clean silicone oil during next minutes.

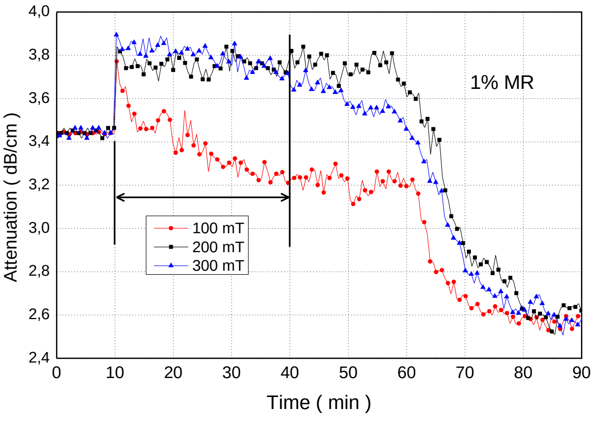

Fig. 3 Experimental data of the changes of acoustic attenuation for a jump change of the magnetic field (● - 100 mT, ■ - 200 mT, ▲ – 300 mT) for 1 wt.% MR fluid measured at 20 °C.

The changes of the acoustic attenuation for the same step changes and time application of the magnetic field for 2 wt.% MR fluid are presented in Fig. 4. Evidently, the quick change of the acoustic attenuation at step change of the magnetic field can be seen again. After an initial decrease of the acoustic attenuation, it did not significantly changed during next time of presence of the magnetic field. When the magnetic field was switched off, a small decrease and increase again was measured. The acoustic attenuation then exponentially decreased with time constant of about 230 s. After this occurrence, the exponential decrease of acoustic attenuation to value as for clean silicone oil can be observed again. For all magnitudes of the magnetic field the development of acoustic attenuation was similar and its value corresponds to the applied magnetic field.

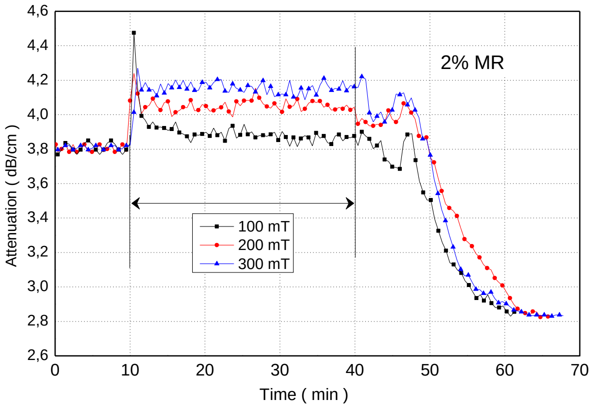

Fig. 4 Experimental data of the changes of acoustic attenuation for a jump change of the magnetic field (■ – 100 mT, ● – 200 mT, ▲ – 300 mT) at for 2 wt.% MR fluid at 20 °C.

Figure 5 illustrates the results of the anisotropy of acoustic attenuation in the magnetic fluid of the value 200 mT studied in the dependence on the angle φ between wave vector $\boldsymbol{k}$ and the magnetic field $\boldsymbol{B}$ measured for various concentration of MR fluids at 20 °C. The results indicate a significant effect of angle on the acoustic attenuation within the fluids. The measured anisotropy for both concentration have similar development, but for 2 wt.% concentration the values of acoustic attenuation are higher. The acoustic attenuation to angle 50 slowly decreases and then increases. There are same minimum between angle 40° and 60°. It is interesting that for parallel orientation of magnetic field and wave vector of acoustic wave the acoustic attenuation was smaller than for perpendicular orientation.

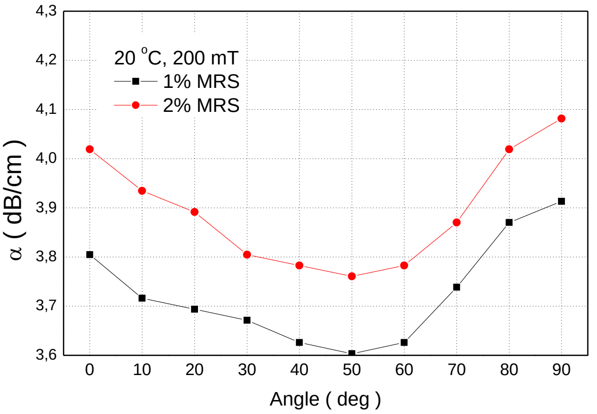

Fig. 5 Anisotropy measurement of the acoustic attenuation at constant magnetic field of 200 mT for 1 wt.% (■), and 2 wt.% (●) MR fluid.

Interesting phenomena was observed when the magnetic field was switched off after anisotropy measurement (Fig. 6). Time of the application of magnetic field during this type of measurement was more than 120 min. In this case for each concentration immediately decrease of acoustic attenuation to value as for clean silicone oil was observed. There was no relaxation time as was observed in Figs. 3 and 4 when the magnetic field was applied only 30 min.

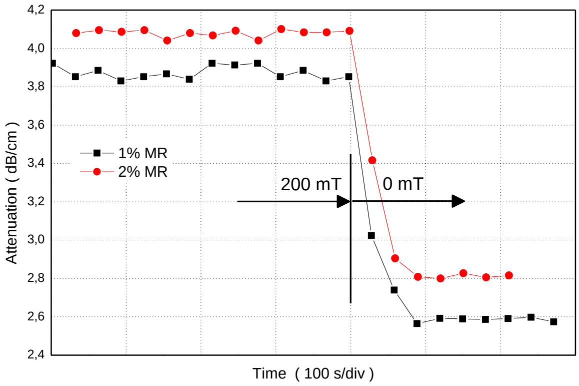

Fig. 6. Experimental data of the changes of acoustic attenuation for a step change of the magnetic field from 200 to 0 mT for various concentration of MR fluids (● – 2 wt.%, ■ – 1 wt.%).

IV. DISCUSSION

It is known that the interaction between the external magnetic field and the magnetic moment of the nanoparticle in FF leads to the aggregation of nanoparticles to new structures (Odenbach 03, Kúdelčík 13, Satoh08, Rozynek 11). These structures enlarge with the magnetic field and this process has the influence on the value of the acoustic attenuation. It can be expected that behavior in MR fluids acoustical characteristic vs. magnetic field will be similar to FF behaviour. So, the acoustic attenuation can be used as important informative parameter characterizing the quality of MR fluids structure.

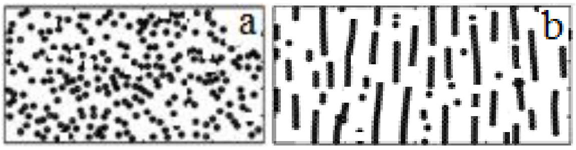

Fig. 7 MR fluid model without (a) and under (b) the magnetic field applied.

Normally, MR fluids are free flowing liquids with chaotic distribution of particles having a consistency similar to that of silicon oil (Fig. 7a). However, in the presence of external magnetic field applied, the CI particles acquire a dipole moment aligned with the field causing the particles form linear chains aligned according to the magnetic field (Fig. 7b). This arrangement was confirmed by other measurements [Baranwall 12, Kciuk 06] where the degree of change is related to the magnitude of the applied magnetic field, and can occur in a few milliseconds. The similar quick effect of the application of magnetic field on the change of acoustic attenuation is shown Figs. 3 and 4. After immediately change of the acoustic attenuation, the next development dependent on the magnitude of applied field and concentration. In the case of 1 wt.% MR fluid at 100 mT the steady decrease could be caused by the effect of acoustic wave, which by the oscillation of particles causing decay of chains. For magnetic field of 300 mT, the chains are so big, that they sediment, what have effect on the acoustic attenuation. The created structures at 200 mT are stable during the whole time of application of magnetic field. In the case of 2 wt.% concentration of CI particles within MR fluid, the acoustic attenuation was almost stable for all magnitudes of magnetic field. This effect may be caused by higher numbers of particles, thus producing more stable chains.

After switching off the magnetic field, three interesting effects were observed. The first of them is long time, i.e. significant changes in acoustic attenuation were noted after several minutes (20 min for 1 wt.% MR fluid and 10 min for 2 wt.% MR fluid). This effect can be connected with different long time of life times of chains. The second is the decrease of acoustic attenuation to the value as for clean silicon oil. It is supposed that this is effect of sedimentation of particles. The process of sedimentation for FF have not been observed since the mass of nanoparticles is much smaller than the mass of microparticles used in MR fluid. Very interesting is the last measurable effect presented in Fig. 6. In this case, the immediate decrease of acoustic attenuation after removing the magnetic field after anisotropy measurements was observed. This may by caused by rapid sedimentation of stable chains created during long period of magnetic field application.

The anisotropy of acoustic attenuation was investigated for two concentrations at20 °C (Fig. 5). As it can be seen from the results there is the significant effect of angle on the acoustic attenuation what is coupled with the process of chains orientation in the direction of field. The chains created from CI magnetic particles are in parallel orientation with wave vector at angle $\varphi = 0°$, so the acoustic attenuation is higher than for other angles. The particles oscillate in the presence of acoustic wave, what causes a loss of the acoustic energy. The increase of interparticles interactions in the chain with concentration had effect on the acoustic attenuation. The acoustic attenuation decreases with increase of φ because at rotation of chains decrease the binding strength between particles influence on the value of acoustic attenuation. This decrease was observed to angle around 50°. The acoustic attenuation is higher for perpendicular orientation as for parallel orientation. This could be caused by the higher possibility of oscillation of the individual particles in perpendicular orientation to the chain, as in the direction of chain. There are also other effect influenced on the acoustic attenuation, like the radius of clusters and the length of their chains. On the basis of the theory for the anisotropy presented in works [Kudelcik 13, Skumiel 04] the influence of translational vibrations and rotational degrees of freedom of chains existed in MR fluids in the presence of magnetic field can be determined.

V. CONCLUSIONS

This study was focused on the magnetic field induced changes of the structure of CI particles in silicon oil based MR fluids. The strong influences of magnetic field on the structures were detected using acoustic spectroscopy. The observed behavior of the acoustic attenuation validated the process of aggregation of the magnetic particles into chain. The exponential decrease of acoustic attenuation to the value for clean silicone oil was observed after switching the magnetic field off. The study of the anisotropy showed the orientation of magnetic field in the MR fluids is also important.

ACKNOWLEDGMENT

This work was supported by projects VEGA 2/0045/13. The authors wish to thank for the support to R&D operational program Centre of excellence of power electronics systems and materials for their components II. No. OPVaV-2009/2.1/02-SORO. The author M. S. would like to thank the Grant Agency of the Czech Republic (14-32114P) for financial support. This work was supported by the Ministry of Education, Youth and Sports of the Czech Republic – Program NPU I (LO1504).

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