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Capillary flow of a suspension of non-magnetic particles in a ferrofluid under highly non-uniform magnetic field

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Abstract

When a ferromagnetic suspension flows through a capillary placed between two small strong permanent magnets, the magnetic force acts upon the non-magnetic (silica) particles dispersed in a ferrofluid and they tend to be extruded from the zone of high magnetic field. Particles get concentrated at the entrance section between magnets and form a plug. The increase of hydraulic resistance is due to the relative motion between particulate and ferrofluid phases in the presence of a field. If we keep the pressure difference constant, the flow rate will decrease when the field is applied and can eventually completely stop. In order to restart the flow a pressure difference, high enough to push the silica plug out of the capillary, is needed. The critical pressure of the flow blockage is nearly two times less than the pressure of the flow onset, both pressures being independent of the particle concentration in the suspension (except for near zero concentration). Such hysteresis of the flow onset/blockage has also been predicted in the frame of the proposed two-phase flow model, which has been used to calculate steady-state concentration profiles and discharge characteristics.

Keywords: Ferrofluid; Magnetorheological suspension; Non-uniform magnetic field; Two-phase flow; Flow blockage

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1. Introduction

Study of a magnetic fluid flow in highly non-uniform magnetic field is of practical interest because of numerous technical and medical applications. The main feature of such a flow is that a non-uniform field causes non-uniform distribution of concentration of magnetic particles in the fluid that, in turn, will change its magnetic and rheological properties. Helpful reviews on hydrodynamics and heat and mass transfer in colloids and suspension of magnetic particles are given in books of Rosensweig (1985) and Blums et al. (1989). Concerning applications, redistribution of the magnetic phase concentration in magnetic fluid seals (Taketomi, 1980) and printers (Maruno et al., 1983) is an undesirable effect. Also, in magnetic fluid separators, after a long operation period the diffusion of ferrofluid nanoparticles can change the characteristics of the device as pointed by Lukashevich et al. (1988). There are however a lot of applications where such diffusion plays a positive role. One of them is the magnetic technique of the separation of biological cells when a so-called magnetic label is attached to chosen cells and the labeled cells are displaced by a magnetic field to the prescribe emplacement (Šafařík and Šafaříková, 1999). Another example is the new blood cancer treatment technique proposed by Flores and Liu (2001). If some micron size magnetic particles are introduced into the blood, they will gather in the domain of high magnetic field and form a seal, which blocks the flow and cut the alimentation in oxygen of a tumor tissue. The authors present in vitro modeling of the blood embolization of the sheep blood. In our paper a new possibility of blood flow blockage using a ferrofluid is discussed.

Once injected to blood, the ferrofluid is supposed to form a homogeneous mixture with blood plasma. Red blood cells, being weakly magnetic compared to this mixture, will behave as magnetic holes in such magnetized surrounding. They will be repulsed by the magnetic field gradient to the domain of weaker field and form a plug, which will block the blood flow. Oppositely to the suspension used by Flores and Liu (2001) where they use micron-sized particles, we do not have to consider a concentration gradient of magnetic particles of the ferrofluid itself in a field gradient, because they are nanometric and Brownian motion prevents them to concentrate in high magnetic field up to several hundreds T/m (see Bashtovoy et al. (1988), Section 2.1).

We should mention here that ferrofluids are nowadays considered as very promising "intelligent" materials for medicine and they have already found some significant applications tested on human being, like magnetic fluid hyperthermia (Jordan et al. (2001)) and magnetic drug targeting (Voltairas et al. (2002)). Note that the use of nanometric size ferrofluid particles instead of micrometric ones seems to be more safe from the viewpoint of the risk of post-therapy thrombosis and their metabolism. Thus, the proposed method of blood embolization could be an alternative to the one of Flores and Liu (2001). The study presented in this paper does not pretend to cover all the biological aspects of the proposed idea; we consider the flow problem from a physical point of view trying to give a qualitative insight of the flow blockage phenomenon. To simulate the red blood cells in the ferrofluid–blood plasma mixture we use a suspension of micron-sized non-magnetic particles dispersed in a ferrofluid and study a flow of this suspension through a capillary with a magnetic field applied on a small part of the capillary.

It is useful now to describe some features of this kind of suspension. The non-magnetic particles act as magnetic holes in a magnetized ferrofluid. By analogy with magnetic particles in a non-magnetic surrounding, they have a magnetic moment. Under applied magnetic field dipole–dipole interaction occurs between non-magnetic particles, they are attracted to each other and form

chain-like aggregates extended along the magnetic field direction. Under uniform magnetic field this suspension behaves in the same way as a classical magnetorheological fluid (suspension of magnetic particles), i.e. have a field-dependent yield stress below which the flow is impossible. Magnetorheology of such a system was investigated by Kashevskii et al. (1988) and Volkova et al. (2000). Under non-uniform magnetic field, the ferrofluid phase of the suspension moves towards the domain of the high field, while non-magnetic particles are repelled and concentrate in the domain of weaker field. This repulsive force can easily compensate the gravity of particles and make them float up. This is in fact a principle of separation of non-magnetic particles by their density described by Khalafalla and Reimers (1973).

Tracking back to the blood embolization we note that it can be realized by the other technique applicable only for large blood vessels (arteries) where it is possible to form a ferrofluid drop thrombus. Khizhenkov et al. (1993) have proposed to inject a tiny volume of ferrofluid into a blood vessel and put a strong permanent magnet near the injection zone in order to hold the ferrofluid drop in the vessel. The authors calculated a sealing capacity of such magnetic thrombus and presented experimental results. Since biocompatible ferrofluids are always mixable with blood, some red blood cells should be present in the blood–ferrofluid mixture. So, one could expect some effect of the non-magnetic red blood cells on the sealing capacity of the magnetic thrombus. But there is no information in literature on this point and we shall try to get it by studying the behavior of a suspension drop inside a capillary.

The behavior of a pure ferrofluid drop in cylindrical channels placed in uniform and non-uniform fields have been studied by Bashtovoi et al. (1987a,b). It has been found that a cylindrical configuration of a non-wetting ferrofluid drop becomes unstable at some critical uniform magnetic field transverse to a capillary, and that the drop transforms to a plane thin film parallel to the field. Such an instability, which, in fact, destroys a drop, should be taken into account in the process of embolus.

In our experiments, statics of the suspension drop and kinetics of a suspension flow blockage are studied. The critical pressure of the flow onset or blockage is measured and a model for the two-phase flow is given that predicts steady-state concentration profiles, discharge characteristics and the critical sealing pressure.

2. Experiments

The experimental setup is shown in Fig. 1a and b. A cylindrical capillary is placed between two cylindrical cobalt-samarium magnets in such a way that the capillary axis crosses at right angle the axis of magnets. Magnets have diameter 13 mm and height 15 mm. A special device allows to displace quickly the magnets along their axes. Thus, the distance between magnets can be changed from 30 cm to the minimal value 1.5 mm when they are in contact with the capillary wall.

Magnetic field induction inside and outside the gap between magnets is measured by Hall gauge. Results of measurements show that inside the gap the magnetic field component, B_z, parallel to the axis of magnets is much larger than the radial component, B_x, (x denotes the coordinate along the capillary axis). The field B_z varies weakly along the width of the gap and strongly along the x direction: $\partial B_z/\partial z \ll \partial B_z/\partial x$. Dependence of the magnetic field induction on the radial coordinate x is presented in Fig. 2. The maximum $(B_z)_{\max} = 0.75\,\mathrm{T}$ of the field induction takes

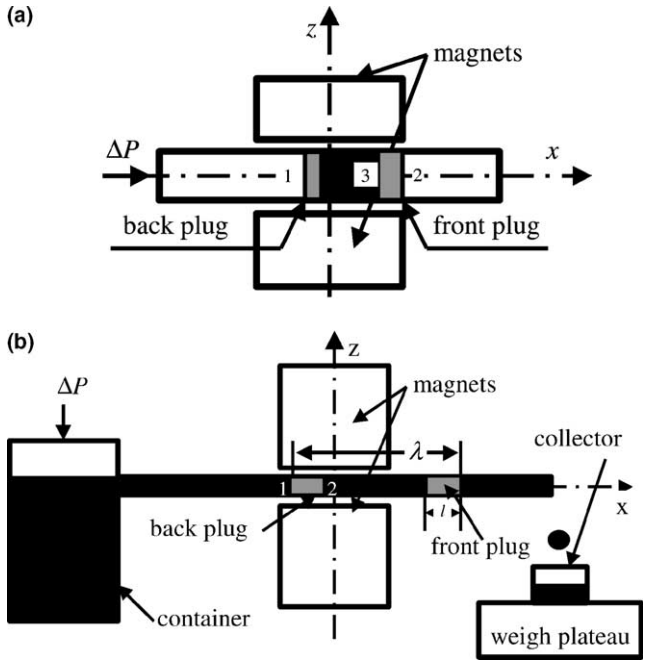

Fig. 1. Experimental setups for investigation of statics of a suspension drop (a) and of suspension flow through a capillary (b).

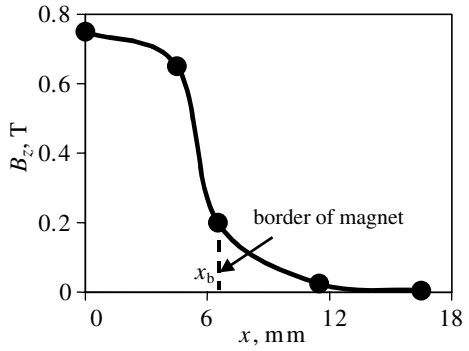

Fig. 2. Distribution of z-component of the magnetic field induction produced by a pair of magnets.

place on the magnets' axis. Outside the gap, the field B_z decreases from 0.2 T on the border $x_b = 6.5$ mm of the gap to 5 mT at 10 mm from the border x_b. We obtained a good fit of the experimental distribution of magnetic field by the following function:

$$g(x) = B_z(x)/(B_z)_{max} = 1/(1 + a \cdot (x/x_{max})^6) \quad (1)$$

with $a = 588$ and $x_{max} = 16.5$ mm being the point where the field is $B_z = 5$ mT. The solid line in Fig. 2 shows this curve.

As cylindrical capillaries, we used PYREX micro-sampling pipettes (Corning Glass Works, USA) made of borosilicate glass. The length of tubes is $L = 127\,\text{mm}$, the volume is 10 ml, that corresponds to internal diameter $2R = 0.32\,\text{mm}$.

The ferrofluid used is a colloidal suspension of magnetite particles in silicon oil 47V10 supplied by Rhodorsil (Lyon, France). Ferrofluid is stabilized by oleic acid and is stable against gravitational sedimentation during several years. Diameter of particles is about 8 nm and mass concentration about 25%. Physical properties are the following: density $\rho_f = 1250\,\text{kg/m}^3$, saturation magnetization $M_f^{sat} = 30\,\text{kA/m}$, initial relative magnetic permeability (permeability at low fields when the magnetization law is linear) $\mu_f^i = 2.7$. Magnetization curve is represented by the Frölich–Kennelly approximation (Bozorth (1951)):

$$M_f(H) = \frac{\mu_f^i - 1}{1 + (\mu_f^i - 1) \cdot H / M_f^{sat}} \cdot H \tag{2}$$

with H being the magnetic field intensity. Viscosity measurements of ferrofluid are performed in a cone-plate geometry of rotational stress-controlled rheometer Haake RS-150. Both in absence and in presence of the magnetic field (up to 30 kA/m) perpendicular to the shear flow, ferrofluid behaves as a Newtonian fluid with the field-independent viscosity $\eta_f = 0.015\,\text{Pa s}$.

We used silica powder particles supplied by Lancaster Synthesis, Pelham, New Hampshire, USA, of mean diameter $D_p = 1.5\,\mu\text{m}$ as non-magnetic inclusions. The density of the particle material is $\rho_s = 2200\,\text{kg/m}^3$. Silica particle dispersion in ferrofluid is performed with the help of an ultrasound tip of capacity 20 W during 15 min. Suspensions of two volume concentrations of silica particles $\Phi_0 = 13\%$ and 30% were made. The experiment is performed just after the preparation of a suspension.

We have studied experimentally two different situations in view of potential application for blood embolism: (1) statics of the suspension drop in a capillary and (2) flow of the suspension through capillary with respect to the redistribution of concentration and seal formation under non-uniform magnetic field.

When studying statics of a suspension drop, a drop of a given volume is placed into a capillary (Fig. 1a). Ferrofluid wets well the capillary, so, the length of the drop is determined as the distance between wetting perimeters and is equal to 6 mm for all drops. Then we approach quickly the two magnets till they are in contact with the capillary in order to obtain the strongest possible field. One of capillary ends is connected with the compressed air cylinder. A micro-valve allows controlling pressure very smoothly. We increase the pressure stepwise until some critical value where the drop darts off and extrudes from the capillary. When approaching the critical pressure we increase the pressure with steps of 100 Pa and wait 10 min before next step.

The second part of experimental studies concerns the continuous flow of the suspension through the capillary. As depicted in Fig. 1b, one capillary end is attached to a container filled with the suspension. The container is connected with a compressed air cylinder, so, the pressure exerted from the compressed air extrudes the suspension from the container to the capillary and thus induces the flow. The non-uniform magnetic field is applied to a short middle section of a capillary by two permanent magnets (Fig. 1b). The applied pressure is always kept constant whatever the flow rate is. As the suspension flows, it drips out from the free capillary end to a collector placed on top of an electronic balance Precisa 40SM-200A, and the mass G of collected fluid is

recorded every 0.5 s. The instantaneous value of the flow rate is calculated from the experimental curve $G(t)$: $Q = (1/\rho)\Delta G(t)/\Delta t$ with ρ being suspension density and t being the time.

The sequence of experimental procedure is as follows. First, we remove the magnets from the capillary in such a way that the magnetic field is zero everywhere along the capillary. Then we apply a certain pressure to the suspension and measure the induced flow rate using the technique indicated above. At last we approach the magnets rapidly to the minimal gap width of 1.5 mm and we begin to record the temporal evolution of the suspension flow rate until some steady-state value is attained. In fact, two flow regimes are observed. The first one is the transitional regime with a decreasing flow rate due to some structural transformations in the suspension after the field application. The second one is the steady-state regime corresponding to a constant flow rate when these transformations have been completed. In experiments we decide that we have reached the steady-state regime if, during several hours, the flow rate decrease does not exceed 5%, which is the uncertainty of measurements. At some external pressures we observed that the flow stopped completely. We have recorded the critical pressure values below which the flow could be stopped by the application of the field. Once the flow had been blocked, we surprisingly had to apply some higher pressure to restart it.

A different study concerned the flow onset after the magnetic field has been applied and held constant during approximately 30 min before applying a pressure. In this case we increased the pressure smoothly departing from zero pressure and recorded the critical value when the flow started.

3. Experimental results

3.1. Statics of a drop (Fig. 1a)

When the magnetic field is applied, the distance between wetting perimeters of the drop increases from 6 to 8 mm, that is related to a change of meniscus configuration (the volume of the drop remains constant). Thus, the whole drop remains in the domain of high magnetic field in the center between magnets. For ferrofluid with silica particles, we can clearly see that two white spots appear at extremities of the drop a few minutes after the magnetic field has been applied (Fig. 1a). These spots indicate domains of high concentration of silica particles (hereinafter we call them plugs). Such a phase separation happens due to magnetic forces, which tend to repel non-magnetic particles outside the high field zone.

As the pressure increases, the suspension drop moves a little towards the capillary outlet and takes its equilibrium position somewhere farther from the center (but always between magnets). At some critical pressure, there is no more equilibrium and the drop leaves the capillary very quickly. It happens when the back meniscus of the drop reaches the magnets' axis. This process has some particularities for the drop of ferrofluid containing silica particles. When the front of the back plug crosses the center, the repulsive magnetic force acting on the front part of non-magnetic plug changes its direction. Therefore silica particles begin to migrate quickly from the back plug to the front plug. Thus, just before the drop darts off, we observe a front white spot much longer and more contrasted than the back one. It indicates that almost all particles have been concentrated near the front meniscus. For drops of same length and three different values of silica particle con-

Table 1
The critical pressure (in kPa) versus the volume fraction (in %) of silica particles in a suspension

Silica concentration Φ_0	Drop		Flow onset		Flow blockage	
	Experiment	Estimation	Experiment	Estimation	Experiment	Calculation
0	14	18	1.6	–	1.0	–
13	16	17	9.3	10	5.5	5.5
30	15	15	9.8	12	5.5	5.3

centration ($\Phi_0 = 0\%$, 13% and 30%) we have obtained approximately the same critical pressure 15 ± 1 kPa (Table 1). Why does this pressure not depend much on the particle concentration Φ_0 and keep nearly the same value even for pure ferrofluid without any silica inclusions? It is probably connected with a strong redistribution of particle concentration within the drop. As already noted, at near critical pressures, the majority of silica particles are collected on the front side of the drop (Fig. 1a). The major part of the drop is now silica free and situated in the zone of the highest magnetic field gradient. Therefore this part of the drop is subject to much higher magnetic force than the front plug. This holding magnetic force, which equilibrates the external pressure, acts mostly on the silica free region of the drop and should be almost the same as for pure ferrofluid and so independent of the concentration of silica particles, Φ_0, initially added to the suspension. But we could expect some effect of the silica concentration Φ_0 for higher values of Φ_0 or shorter drops when front silica plug would extend to high field gradient region. A quantitative estimation of the critical pressure is given in Section 4.1.

Finally note that a cylindrical shape of the suspension drop remains stable for any equilibrium position of a drop between magnets. Similarly to the case of non-wetting ferrofluid drop studied by Bashtovoi et al. (1987a), the highly non-uniform magnetic field near menisci prevents the drop from extension along the capillary axis, that would happen in uniform field.

3.2. Capillary flow (Fig. 1b)

Let us consider first the onset of the suspension flow through the capillary when the magnetic field is applied a long time before the pressure is. As soon as the field is applied to the magnetic suspension at rest, the silica particles are extruded from the magnetic field region between magnets towards each side of this region. A few seconds later, we observe two plugs of silica particles located symmetrically relative to the magnets' axis at a distance of 4–5 mm from the lateral face of magnets, i.e. in domains of low magnetic field. The plugs' length is about 4–5 mm for the suspension of silica concentration $\Phi_0 = 0.13$ and about 12 mm for the silica concentration $\Phi_0 = 0.3$. When the pressure is applied these plugs move and one of them (back plug) takes an equilibrium position closer to magnets' axis and the other one (front plug) farther. The force exerted by the applied pressure equilibrates the magnetic repulsive force acting on the back silica plug and the flow does not occur. We have not observed a change of the plugs' length while displacing along the capillary. As soon as the back plug front crosses the central line, the maximal magnetic force (holding the back plug) is achieved and the whole plug is quickly expelled from the magnet's area and the flow starts. This flow extrudes both plugs from the capillary. In the same manner as for the drop, the critical pressure when the flow starts is found to be almost the same for the

two volume fractions of silica initially added to the suspension: $\Phi_0 = 13\%$ and 30% and roughly equal to 10 kPa (Table 1). This is because the repulsive force is proportional to the volume fraction Φ_s of particles forming the back plug, but not to the initial volume fraction Φ_0 of the homogeneous suspension. The concentration Φ_s corresponds roughly to a random packing of the particles in the plug and does not depend on the initial concentration Φ_0 of the suspension, except for near zero concentrations when there is simply not enough particles to form the plug.

In this last case, namely in pure ferrofluid, we could nevertheless observe some "blocking" effect. If the pressure is applied immediately after the application of the field, flow occurs at pressures larger than 0.6 kPa, if we applied it two hours after the field we obtain a critical pressure $\Delta P = 1.6$ kPa. Since the process of magnetic sedimentation in ferrofluids is quite slow (see Lukashevich et al. (1988)), the result $\Delta P = 0.6$ kPa is supposed to be due to non-Newtonian yielding behavior of the fluid because of structuring in high magnetic fields (even though the ferrofluid remains Newtonian in weak fields up to 30 kA/m). The observation of an increase of the critical pressure with time can be due to both redistribution of magnetite particle concentration in highly non-uniform field and to an increase of the yield stress because of long-lasting process of structure formation.

We are now coming to the most important part of the experimental study—flow deceleration after magnetic field application. This is the case of interest, if we want to stop the blood flow in capillaries. In experiments, the field was applied after the flow had already been induced by the external pressure difference. We observe that, below some critical pressure the flow is completely stopped by the magnetic field. During flow deceleration, we could see a white spot forming in the capillary just before the magnets. As the flow rate decreased, this spot (corresponding to the back plug) became brighter and increased in length. After the flow had stopped the silica particles that had not still left the magnetic field zone between the magnets moved away to domains of weaker magnetic field. If the particles were located between the back plug and the center of magnets, they moved towards the back plug; if they were located on the other side of the magnets' center, they moved towards the capillary outlet and formed a front plug (Fig. 1b). According to our observations, the front plug was visually shorter than the back one. After the complete stop of the flow, the plugs were not symmetric relative to the magnets' axis. The back one was located closer to this axis and, at near critical pressure, was standing between magnets (Fig. 1b). The front plug was located at a distance of 4–5 mm from the lateral side of magnets, i.e. in the domain of weak field, so, the applied pressure was mainly balanced by the magnetic force which tends to expel the back plug towards the capillary inlet.

The critical pressure of the flow blockage was almost the same for both particle volume fraction: $\Phi_0 = 13\%$ and 30% and equal to 5.5 kPa (Table 1). This pressure is near two times less than the critical pressure in case of the flow onset, i.e. when the field is applied first well before the application of pressure. This discrepancy cannot be explained from the simple consideration of the balance of forces acting upon a plug. We shall explain it in the following section where we take into account kinetics of redistribution of the silica concentration in the suspension flow.

The critical pressure of the blockage of pure ferrofluid flow is about 1 kPa that is to say about 60% less than the corresponding pressure for a magnetic field applied first, well before the pressure.

At supercritical pressure the flow does not stop and we no longer see the plugs clearly. Time-dependence of the flow rate for different pressures is shown in Fig. 3a for pure ferrofluid and in

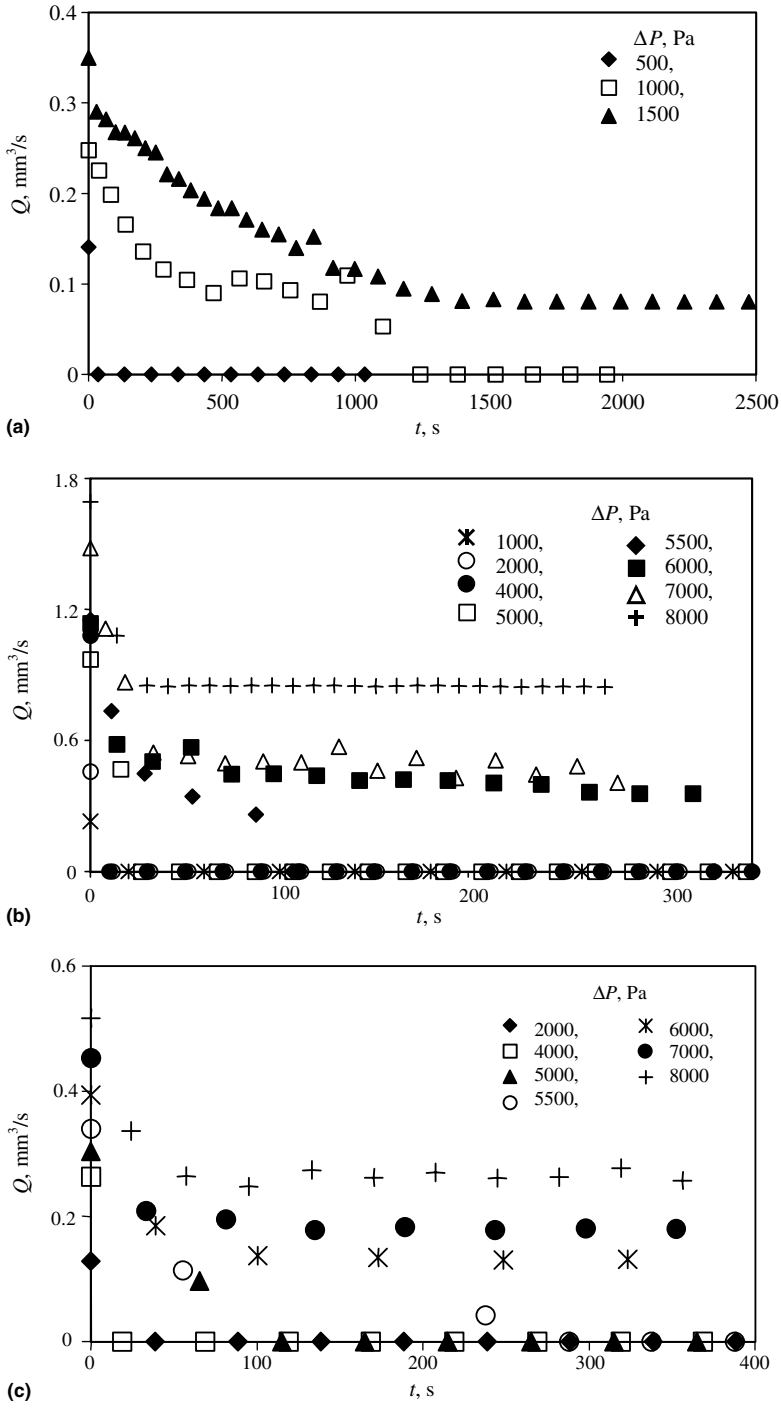

Fig. 3. Time dependence of the flow rate in a capillary for pure ferrofluid (a), and suspension of silica particles in ferrofluid with particle concentration $\Phi_0 = 13\%$ (b) and 30% (c).

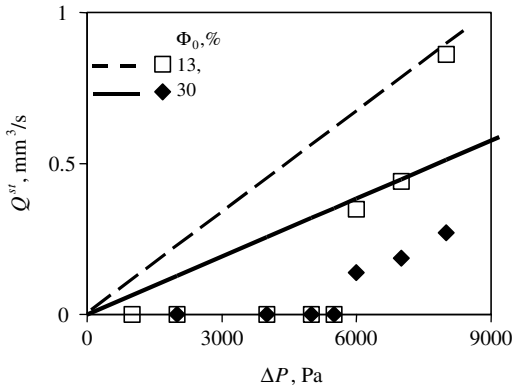

Fig. 4. Dependence of the steady value of the flow rate versus the applied pressure. Lines correspond to the flow rate in the absence of the magnetic field.

Fig. 3b and c for ferrofluids with silica particles of volume fraction 13% and 30% correspondingly. All the three figures have the same appearance. Within a wide range of subcritical pressures, the flow stops almost immediately (drops stop to fall into collector just after the sudden approach of the magnets near the capillary, thus, the sealing time is a few seconds). A long-time blockage process can be observed only at pressures close to the critical one. At supercritical pressures, the flow rate decreases up to some steady value Q^{st} after a lapse of time. This time is about 0.5–2 min for ferrofluid with silica particles and 0.5 h for pure ferrofluid. Dependence of the steady flow rate Q^{st} on the applied pressure ΔP is shown in Fig. 4 for a ferrofluid with particles (points). For pressure up to 8 kPa, Q^{st} is still much smaller than the initial flow rate Q_0 in the absence of the field (lines). Such a strong decrease of the flow rate is explained first of all by the friction between silica particles and ferrofluid due to a large difference between velocities of the particulate and ferrofluid phases. The other reason can be connected with yielding rheological behavior of a suspension of non-magnetic particles in a magnetic field. Which mechanism is predominant? Necessary estimations are given in Section 4.2. From these estimations we have built a simple two-phase flow model, which will allow us to make some predictions concerning critical pressure and the steady-state flow.

Finally note that linearity of the flow rate Q_0 on the pressure ΔP shows Newtonian behavior of suspensions in the absence of magnetic field. Thus, the flow rate versus the pressure is represented by the Poiseuille formula:

$$Q_0 = \frac{\pi R^4}{8\eta L} \cdot \Delta P, \tag{3}$$

where the suspension viscosity η is expressed through the viscosity η_f of ferrofluid and volume fraction Φ_0 of spherical silica particles by Wand formula (Bashtovoy et al. (1988), Section 2.1):

$$\eta = \eta_f \cdot \exp\left(\frac{2.5\Phi_0 - 2.7\Phi_0^2}{1 - 0.609\Phi_0}\right). \tag{4}$$

We found a good agreement between the formula (3) and experimental discharge curves $Q_0(\Delta P)$ in the absence of field.

4. Theory and discussion

4.1. Critical pressure estimations

In this section we shall estimate critical sealing pressure in the static cases (the drop and onset of the flow) where we do not have to consider kinetics of the seal formation. The pressure in question will be found from the balance between the external pressure and the magnetostatic pressure in the suspension.

According to Taktarov (1980), the gradient of the mean magnetostatic pressure in the suspension is defined through the mean ferrofluid magnetization M_f and the gradient of the mean magnetic field intensity H_f in the ferrofluid phase:

$$\nabla P = \Phi_f \mu_0 M_f \nabla H_f, \tag{5}$$

with Φ_f being the volume fraction of ferrofluid phase and μ_0—the magnetic permeability of vacuum.

Thus, to obtain the external pressure difference ΔP, we just have to integrate the Eq. (5) either over the length of the suspension drop or, in the case of the flow onset, over the length of the whole capillary containing suspension:

$$\Delta P = \mu_0 \int \Phi_f M_f \, dH_f. \tag{6}$$

In the case of a pure ferrofluid we have $\Phi_f = 1$ and recover the well-known formula (Rosensweig (1985), Section 4.2) $\Delta P = \mu_0 \int M_f dH_f$ for the pressure difference. For a pure ferrofluid drop this formula gives a critical pressure value defined by the difference in magnetic field values between the back (H_{f1}) and the front (H_{f2}) menisci of the drop. For the pure ferrofluid filling the whole capillary the difference in magnetostatic pressure on the capillary inlet and outlet is zero because the magnetic field is zero at these locations. So, we should not have any yield pressure for the onset of pure ferrofluid flow. This is not the case in experiments because, as previously discussed, we have some long-lasting magnetic sedimentation in the ferrofluid when its nanoparticles migrate towards magnets. However this process is very slow compared to the formation of the silica plug in the suspension and experimental value $\Delta P = 1.6\,\text{kPa}$ for the onset of the ferrofluid flow is one order of magnitude lower than the critical value for the onset of the silica suspension flow. Thus, it is justified to neglect the magnetic sedimentation effect in our estimations.

In the case of silica suspension the integral (6) should be taken separately for the domains containing silica particles and those free of particles. In case of a drop of suspension close to critical pressure, we have a pronounced front plug with some packed concentration Φ_s^{pack} of silica and a pure ferrofluid zone extending from the back meniscus to the front plug (the back plug has already disappeared because of silica migration towards the front plug). The critical pressure difference takes the following form for the drop:

$$\Delta P = -\mu_0 \int_{H_{f1}}^{H_{f3}} M_f \, dH_f - \mu_0 \left(1 - \Phi_s^{\text{pack}}\right) \int_{H_{f3}}^{H_{f2}} M_f \, dH_f. \tag{7}$$

Here H_{f3} is the magnetic field strength in the ferrofluid on the left border of the front plug (cf. point 3 in Fig. 1a).

In the case where the whole capillary is filled with the suspension (cf. Fig. 1b), we have to consider three different zones. The first one extends from the capillary inlet to the back plug, being subject to relatively weak magnetic field varying from zero at the inlet to H_{f1} at the left side of the back plug. The silica concentration in this zone is supposed to be quasi-uniform and equal to Φ_0. The second zone is the back plug with some pack silica concentration Φ_s^{pack}, and the third one is silica free zone ($\Phi_f = 1$) between the back and front plugs where magnetic field varies from H_{f2} on the right side of the back plug to zero. The front plug and the zone between this plug and the capillary outlet are subjected to zero (or, more precisely, negligibly weak) magnetic field and are not taken into account in the integration of (6). This integration gives us

$$\Delta P = -\mu_0(1-\Phi_0)\int_0^{H_{f1}} M_f\,dH_f - \mu_0\left(1-\Phi_s^{pack}\right)\int_{H_{f1}}^{H_{f2}} M_f\,dH_f - \mu_0\int_{H_{f2}}^{0} M_f\,dH_f. \tag{8}$$

To exploit the obtained formulas (7) and (8) we need to determine the mean magnetic field H_f in the ferrofluid phase of the suspension and the mean magnetization M_f in the ferrofluid phase, both intervening into these expressions. The magnetization vs. field strength dependence is given by Eq. (2). Analysis of this expression shows that, at the magnetic field higher than 0.1 T, the ferrofluid magnetization is near its saturation: $M_f \approx M_f^{sat}$. Furthermore, even if we use the approximation $M_f \approx M_f^{sat}$ for the whole magnetic field range along the capillary 0–0.75 T, we will have less than 7% error in ΔP compared to precise calculations using nonlinear magnetization law (2). Concerning the mean magnetic field H_f in the ferrofluid phase of the suspension, in general, it is determined through the mean magnetic permeability of the suspension, which depends, on the volume fraction of solid phase, shape of the particles and their arrangement in the suspension. Numerous mean field theories exist to treat this problem, a helpful review given in the book of Berthier (1993). However, if the external magnetic field H is high enough compared to the ferrofluid saturation magnetization M_f^{sat}, the mean magnetic field H_f will be close to the external one, the difference $H - H_f$ being always less than M_f^{sat} (see, for instance, Taktarov (1980)). The main contribution to the pressure difference ΔP is given within the field range 0.1–0.75 T, and it is not surprising that we may use the approximation $H_f \approx H$ for the whole range 0–0.75 T with 3% error in ΔP compared to more exact Maxwell–Garnett formula (Berthier (1993), Section 8.3). Using both approximations $M_f \approx M_f^{sat}$ and $H_f \approx H$, we derive the final expressions, respectively (9) for the critical pressure of the drop and (10) for that of the flow onset:

$$\Delta P = \mu_0 M_f^{sat}\left[H_1 - H_2 - \Phi_s^{pack}\cdot(H_3 - H_2)\right], \tag{9}$$

$$\Delta P = \mu_0 M_f^{sat}\left[\Phi_s^{pack}(H_2 - H_1) + \Phi_0 H_1\right]. \tag{10}$$

In these equations H_1, H_2 and H_3 are the values of the external magnetic field taken in points 1, 2 and 3, correspondingly (cf. Fig. 1a and b).

To calculate ΔP numerically, we only need to determine the value of silica concentration Φ_s^{pack} in the plugs. Quick estimations of Φ_s^{pack} can be done using experimental approximate values of the plugs' length l. This length was measured at zero external pressure when both plugs had equal length. In both cases: drop or a suspension filling the whole capillary, we use the conservation of the particulate phase volume before and after application of the magnetic field. Let λ be the

length of the drop or distance between the outer sides of plugs (Fig. 1b). Then the conservation of silica quantity reads: $\Phi_0\lambda = \Phi_s^{\text{pack}} \cdot 2l$. Hence, $\Phi_s^{\text{pack}} = \Phi_0\lambda/(2l)$. In drops, the plugs' length was near 1 mm for the suspension of silica concentration $\Phi_0 = 0.13$ and near 2 mm for $\Phi_0 = 0.3$. The plugs' length in the case of suspension filling the whole capillary was 4–5 mm for the suspension of $\Phi_0 = 0.13$ and 12 mm for $\Phi_0 = 0.3$, the distance between the inner sides of plugs was 21–23 mm. Thus, for all the plugs, we obtain the packed concentration Φ_s^{pack} between 0.5 and 0.6 that indicates that the particle arrangement inside the plugs is little bit more dilute than in densely packed arrays of spheres.

Finally, since we know the position of the plugs at critical pressure as well as their length, we estimate the values H_1, H_2, H_3 of the external magnetic field corresponding to plug's positions in the capillary by using experimental field distribution (Fig. 2, Eq. (1)). Taking the value $\Phi_s^{\text{pack}} = 0.55$, we obtain the following critical pressures for the drop: 18 kPa for $\Phi_0 = 0$; 17 kPa for $\Phi_0 = 0.13$; 15 kPa for $\Phi_0 = 0.3$ and the following values for the flow onset: 10 kPa for $\Phi_0 = 0.13$; 12 kPa for $\Phi_0 = 0.3$. These theoretical values together with the measured ones are given in Table 1. We see a quite good correspondence between experiments and estimations, the latter confirming that the critical pressure does not depend much on the silica concentration Φ_0 in the suspension (except, of course, for zero concentration in the case of flow onset) for the reasons discussed in Section 3.

4.2. Two-phase flow model

In this section we shall develop a two-phase model of the suspension flow through a capillary under non-uniform magnetic field. This model takes into account temporal evolution of the particles' concentration field since the moment of the field application. The particular case of the steady-state flow, after the concentration field no more evolves with time, will be treated numerically, that will allow us to predict the critical blockage pressure and to explain why it is smaller than the onset flow pressure.

We recall that there are two magnetic field effects responsible for high energy dissipation in the suspension flow. First, the non-uniform field induces migration of non-magnetic particles towards the weaker fields and Stokes friction forces occur between particles and ferrofluid due to their relative motion. Second, the magnetic field induces formation of aggregates, they can span the whole cross-section of the capillary and block the flow. So, below some yield pressure difference the flow does not occur. Let us estimate whether this yielding rheological behavior of the suspension could contribute a lot to the pressure difference required to push the suspension through the capillary. To estimate the yield viscous stress we shall use the chain model of magnetorheological suspension in uniform magnetic field developed by Volkova et al. (2000):

$$\tau_Y = (9\Phi_0/8) \cdot [\tan(\theta_c)/(1 + \tan^2(\theta_c))^2] \cdot \mu_0\mu_f(H)\beta_m^2 H^2, \tag{11}$$

where θ_c is the angle of the chain inclination relative to the magnetic field direction, $\tan(\theta_c) = 2/\sqrt{5}$ in dipole–dipole approach, $\beta_m = (1 - \mu_f)/(2 + \mu_f)$. In high magnetic fields $H \gg M_f^{\text{sat}}$, the magnetic permeability of a ferrofluid is $\mu_f(H) = 1 + M_f^{\text{sat}}/H$ and $\beta_m = -M_f^{\text{sat}}/(3H)$. So, taking the limit $M_f^{\text{sat}}/H \to 0$, we obtain an asymptotic expression for the yield stress in infinite magnetic field:

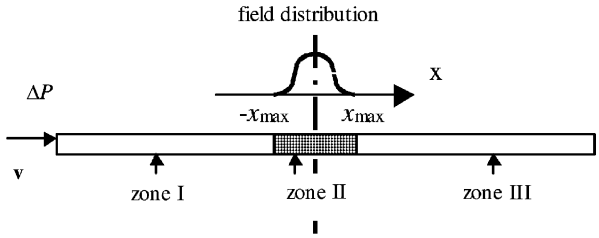

Fig. 5. The different zones of flow. In zones I and III the volume fraction of particles is constant. In zone III the volume fraction and the field depend on position, x.

$$\tau_Y = \left(5\sqrt{5}\Phi_0/324\right) \cdot \mu_0 \left(M_{\mathrm{f}}^{\mathrm{sat}}\right)^2. \tag{12}$$

Thus, we obtain $\tau_Y \approx 5\,\mathrm{Pa}$ for $\Phi_0 = 13\%$ and $\tau_Y \approx 12\,\mathrm{Pa}$ for $\Phi_0 = 30\%$. Estimation of the pressure loss along the section of the capillary subjected to high magnetic field (the length of this section being $l_Y \approx 10\,\mathrm{mm}$) gives $\Delta P_Y = 2\tau_Y l_Y/R \approx 0.7\,\mathrm{kPa}$ for $\Phi_0 = 13\%$ and $\Delta P_Y \approx 1.5\,\mathrm{kPa}$ for $\Phi_0 = 30\%$. It means that the yield behavior of the suspension (accompanied by an abrupt viscosity growth in the field) does not play the major role in the blockage of the flow and will not be taken into account in the model.

To get started with the model let us make the following assumptions:

(1) We divide the capillary into three sections, as shown in Fig. 5. In each section, only one of the two viscous forces is supposed to act: either friction Stokes force (in the middle zone II, where the magnetic field is applied) or shearing force (in zones I, III, where the magnetic field is negligibly weak). Width of the middle zone II is defined by the points $x_{\max} = \pm 16.5\,\mathrm{mm}$ where the magnetic field is $B(x_{\max}) = 5\,\mathrm{mT}$, i.e. negligible relative to the maximal value $B_{\max} = 0.75\,\mathrm{T}$.

(2) In the 1st and the 3rd zones, with no magnetic field applied, the concentration of particles remains homogeneous and equal to Φ_0. Thus the suspension is considered as a single-phase fluid with its viscosity given by Eq. (4) as function of the particle concentration. The pressure loss over the length L_i of the ith zone ($i = \mathrm{I}, \mathrm{III}$) is given by Poiseuille formula

$$\Delta P_i = (8\eta(\Phi_0)L_i/R^2) \cdot v, \tag{13}$$

with $v = Q/(\pi R^2)$ being the suspension velocity.

(3) In the middle zone, aggregates of non-magnetic particles are formed due to magnetic field induced dipole–dipole interactions. The magnetic field gradient induces relative motion of these aggregates relative to the surrounding ferrofluid. Thus, in this zone, our suspension presents two-phase medium with aggregate phase having the volume fraction Φ_{a} and ferrofluid phase comprising only the ferrofluid between the aggregates (but not inside them) and having the volume fraction Φ_{f}. So, $\Phi_{\mathrm{a}} + \Phi_{\mathrm{f}} = 1$. The following assumptions are made with respect to the aggregates:

(3.1) They have all spherical shape, the same size (radius R_{a}) and the same internal volume fraction ϕ of silica. The concentration ϕ is taken to be 0.64 corresponding to a porous medium formed by disordered packing of spherical particles, and the radius R_{a} is the unknown parameter of our system.

(3.2) All the silica free space of the aggregates is filled with the ferrofluid. Ferrofluid motion inside the porous aggregates is not considered, only the flows between aggregates are believed to affect the Stokes friction force acting on the solid phase of the suspension.

(3.3) Kinetics of the particles' aggregation when they enter into the zone of high field is not considered. However, the model takes into account the kinetics of redistribution of the concentration of aggregates, which seems to be the main effect responsible for the flow blockage.

(4) While treating the two-phase flow problem, we make the following assumptions concerning the forces intervening in the momentum equations:

(4.1) Inertial forces and virtual mass effects are neglected because they are quite small compared to Stokes friction forces: the Reynolds number calculated for the aggregates' radius $R_a = 20\,\mu m$ is $Re = \rho_f v R_a/\eta_f \sim 0.02 \ll 1$. We suppose the flow to be laminar both in the macroscopic and in the microscopic (around the aggregates) scales.

(4.2) We neglect both Newtonian and yield part (coming from the magnetorheological effect) of viscous shear forces of the Bingham magnetic suspension. The ratio of the Newtonian part of the shear force density ($\sim\eta_f v/R^2$) to the Stokes force density ($\sim \eta_f v/R_a^2$) is about 0.02. The smallness of the yield shear force has been demonstrated in the beginning of this section.

(4.3) Gravitation forces ($\sim(\rho_s - \rho_f)g$) are negligible compared to magnetic ones ($\sim \mu_0 M_f^{sat}|\nabla H|$) and are not taken into account.

(4.4) We assume that there is no difference between volume average pressure and interfacial average pressure. As indicated by Drew (1983), this assumption is used if the speed of sound in each phase is large compared with velocities of interest.

(5) Magnetic field effect on such flow appears through the magnetic forces $\mathbf{f}_{mf} = \Phi_f \mu_0 M_f \nabla H_f$ and $\mathbf{f}_{ma} = \Phi_a \cdot (1-\phi) \cdot \mu_0 M_{fa} \nabla H_{fa}$ acting on the ferrofluid and aggregate phase of the suspension respectively. The former expression has been obtained by Taktarov (1980) using the volume averaging procedure. The latter force acts upon the silica free ferrofluid part of aggregates having the volume fraction $1-\phi$. M_f and H_f are mean magnetization and mean magnetic field intensity in the ferrofluid phase between aggregates. M_{fa} and H_{fa} are the corresponding values in the ferrofluid phase inside porous aggregates. For the magnetic properties, we shall use the approximation $M_f \approx M_{fa} \approx M_f^{sat}$ and $H_f \approx H_{fa} \approx H$ in the whole second zone, the validity of the approximation being proved in Section 4.1.

To describe the flow in the middle section II we use classical averaged transport equations of two-phase medium formulated by Drew (1983) for a suspension of solid particles in a fluid. We just have to add magnetic forces in the momentum equations and then the system of equations will take the following form according to the above assumptions:

$$-\Phi_f \nabla P_f - \Phi_f \beta(\mathbf{v}_f - \mathbf{v}_a) + \Phi_f \mu_0 M_f^{sat} \nabla H = 0, \tag{14}$$

$$-\Phi_a \nabla P_a + \Phi_f \beta(\mathbf{v}_f - \mathbf{v}_a) + \Phi_a \cdot (1-\phi) \cdot \mu_0 M_f^{sat} \nabla H = 0, \tag{15}$$

$$\partial \Phi_f / \partial t + \nabla \cdot (\Phi_f \mathbf{v}_f) = 0, \tag{16}$$

$$\partial \Phi_a/\partial t + \nabla \cdot (\Phi_a \mathbf{v}_a) = 0, \tag{17}$$

$$\Phi_f + \Phi_a = 1. \tag{18}$$

For all the quantities in these equations subscripts a and f stand respectively for aggregates' phase and ferrofluid's phase between aggregates. The second term of the momentum equations (14) and (15) represents the Stokes force of friction between aggregates and surrounding ferrofluid moving with different mean velocities $\mathbf{v}_a$ and $\mathbf{v}_f$. The friction coefficient β is independent of the velocity for the pure Stokes flow, but is a function of the aggregates' phase concentration $\beta(\Phi_a)$. For this function we apply the results of Zick and Homsy (1982) of numerical simulations of Stokes flow past a periodic array of spheres. In term of the dimensionless drag coefficient $K(\Phi_a)$, introduced by Zick and Homsy (1982), the expression for β reads:

$$\beta = (9/2) \cdot \left(\eta_f/R_a^2\right) \cdot \Phi_a(1 - \Phi_a)K(\Phi_a). \tag{19}$$

The formula (19) with the coefficient $K(\Phi_a)$ calculated for the face central cubic (FCC) arrangement of spheres well recovers known results at small and high (near packing) concentrations of particles even if they are randomly distributed. We have approximated the tabulated function $K(\Phi_a)$ by the exponent $K(\Phi_a) = \exp(b \cdot \Phi_a)$ with the free parameter $b = 8.08$.

To close the system (14)–(18) we need the relation between the pressures P_f and P_a. Following Drew (1983), the most common practice is to put these pressures equal to each other if there is no contact between solid particles of a suspension. We introduce this relation with the limitation for the aggregate concentration $\Phi_a < 0.64$ corresponding to contact of aggregates:

$$P_f = P_a = P \quad \text{if } \Phi_a < 0.64. \tag{20}$$

In case of contact P_f is no longer equal to P_a and we close the system (14)–(18) by fixing the concentration $\Phi_a = 0.64$.

The first two equations (14) and (15) give the pressure gradient as well as the relation between velocities:

$$\nabla P = (1 - \Phi_a \phi) \cdot \mu_0 M_f^{sat} \nabla H = f(x), \tag{21}$$

$$\mathbf{v}_f - \mathbf{v}_a = (\Phi_a \phi/\beta) \cdot \mu_0 M_f^{sat} \nabla H. \tag{22}$$

The second pair of Eqs. (16) and (17) gives the following relation:

$$\Phi_f \mathbf{v}_f + \Phi_a \mathbf{v}_a = \mathbf{v} = Q(t)/(\pi R^2) \cdot \mathbf{i}_x, \tag{23}$$

with $\mathbf{v} = \mathbf{v}(t) = Q(t)/(\pi R^2) \cdot \mathbf{i}_x$ being the superficial velocity defined by the flow rate Q, $\mathbf{i}_x$ being the unit vector in flow direction.

Using Eq. (22) and the first equality of (23), we express velocities $\mathbf{v}_f$ and $\mathbf{v}_a$ through known quantities and substitute them inside Eq. (17). Finally we obtain the following equation for the time-dependent concentration profile of our suspension:

$$\frac{\partial \Phi_a}{\partial t} + \nabla \cdot \left[\Phi_a \mathbf{v} - \frac{\Phi_a^2(1 - \Phi_a)\phi}{\beta} \cdot \mu_0 M_f^{sat} \nabla H \right] = 0. \tag{24}$$

The pressure difference ΔP_{II} over the length of the middle section is defined by integrating Eq. (21). The pressure loss in this zone due to viscous shearing forces can be evaluated by the Poiseuille formula (Eq. (13)) with the suspension viscosity taken for the initial concentration Φ_0 of silica particles. Thus, summation of the pressure losses over the three sections of the capillary will give the following discharge curve:

$$\Delta P = (8\eta(\Phi_0)L/R^2)\cdot v - \mu_0 M_f^{sat}\int_{-x_{max}}^{x_{max}}(1-\Phi_a\phi)\cdot(dH/dx)\,dx. \tag{25}$$

In the latter expression we have taken into account that the magnetic field depends only on axial coordinate x of the capillary.

An analytical solution for the concentration profile can be obtained for the steady flow at $t\to\infty$ and $\partial\Phi_a/\partial t = 0$. For this case Eq. (24) for the concentration profile, Φ_a, is solved with the boundary condition: $\Phi_a(\pm x_{max}) = \Phi_0/\phi$. This condition denotes that the concentration $\Phi_a(\pm x_{max})$ of aggregates on the borders of the middle section corresponds to the volume fraction Φ_0 of particles initially added to the suspension. We get the following solution:

$$\Phi_a\cdot v - \frac{\Phi_a^2(1-\Phi_a)\phi}{\beta}\cdot\mu_0 M_f^{sat}\frac{dH}{dx} = \frac{\Phi_0}{\phi}\cdot v. \tag{26}$$

Introducing the scales $[x] = x_{max}$ for the distance and $[H] = H_{max}$ for the magnetic field intensity and using Eq. (19) for β, we obtain the dimensionless solution of this problem:

$$\frac{\Phi_0}{\phi\cdot\Phi_a(x)} = 1 - \frac{\phi\cdot g'(x)}{A\cdot K(\Phi_a)}, \tag{27}$$

where $A = (9\eta_f x_{max}\cdot v)/(2R_a^2\mu_0 M_f^{sat}H_{max})$ is the parameter characterizing Stokes—to magnetic force ratio, $g'(x) = -6ax^5/(1+ax^6)^2$ is the gradient of the dimensionless field distribution $g(x)$ defined by Eq. (1).

Eq. (27) is solved numerically for two initial concentrations $\Phi_0 = 0.13$ and 0.3 and various values of A. The results are presented in Fig. 6a and b. We see that the concentration profile is asym-

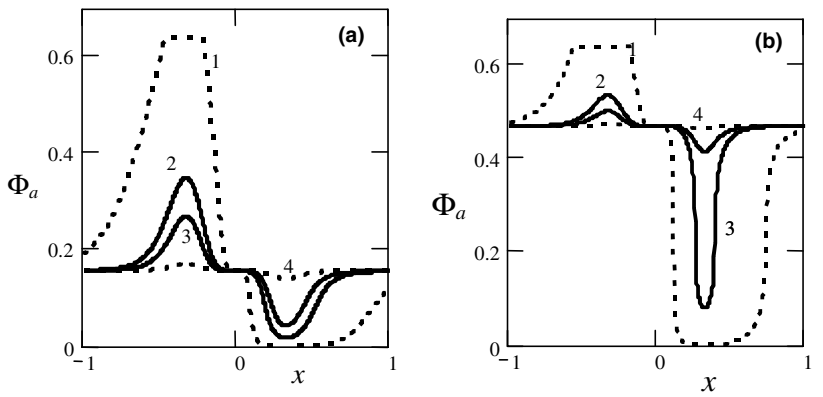

Fig. 6. Concentration profile for initial concentrations $\Phi_0 = 0.13$ (a) and $\Phi_0 = 0.3$ (b) and different values of parameter A: (1) 0.0077, (2) 0.31, (3) 0.77, (4) 7.7.

metric about the origin $x = 0$. It follows from the fact that aggregates are repelled by the magnetic field when they arrive in the high field zone (Fig. 1b); so they slow down and concentrate in the left-hand part of the capillary. Having passed the region of the maximal field they are pushed away by the magnetic field, so the concentration in the right-hand part of the capillary decreases well below the initial volume fraction. At high velocity of the suspension (high values of A), the aggregates (or particles) do not have enough time to get concentrated, they pass through the magnetic field zone practically without changing their initial concentration (that is to say Φ_0/ϕ, curves 4 in Fig. 6a and b). At low values of A, i.e. at low velocities, all the aggregates tend to be concentrated before the center and to be repulsed from the right side of the middle section II. The asymptotic solution for $A = 0$ brings us to the situation of the plug formation in the left part of the second zone ($\Phi_a = 0.64$ at $-1 < x < 0$) with the right part being silica free ($\Phi_a = 0$ at $0 < x < 1$). Here we keep in mind that the coordinate x is normalized by the half length $x_{max} = 16.5\,\text{mm}$ of the middle section II (cf. Fig. 5).

Substituting the solution (27) for Φ_a into Eq. (25), and normalizing ΔP by the magnetic pressure $\mu_0 M_f^{sat} H_{max}$ (the dimensionless pressure being $p = \Delta P/(\mu_0 M_f^{sat} H_{max})$), we obtain the dimensionless discharge curve $p(A)$:

$$p(A) = \frac{16}{9} \cdot \left(\frac{R_a}{R}\right)^2 \cdot \left(\frac{\eta(\Phi_0)}{\eta_f}\right) \cdot \left(\frac{L}{x_{max}}\right) \cdot A - \int_{-1}^{1} (1 - \Phi_a \phi) g'(x)\,dx. \tag{28}$$

Theoretical and experimental curves $p(A)$ are presented in Fig. 7. Lines 1 and 2 correspond to the steady-state flow under magnetic field and 3, 4—to the Poiseuille flow in the absence of field. To obtain the better agreement between theory and experiment, the only parameter of our model—the radius of aggregates of silica particles—is taken as $R_a = 20\,\mu\text{m}$. We see that the func-

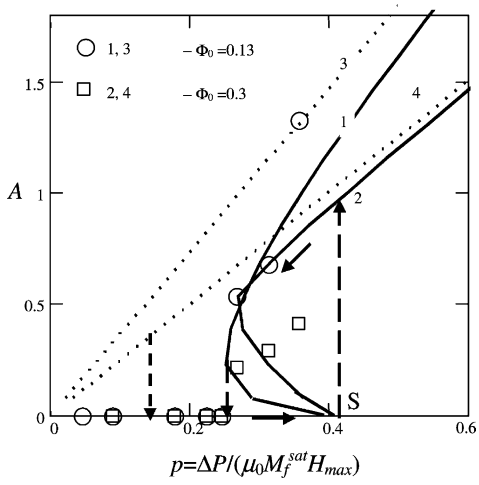

Fig. 7. Dimensionless discharge curves for the steady flow in the presence of the magnetic field (solid: theoretical curves) compared to experimental points. The dashed curves are Poiseuille flow in the absence of the magnetic field. Left vertical arrow illustrates temporal evolution of the flow rate since the moment of the field application. Other arrows illustrate a hysteresis loop of the discharge curve.

tion $p(A)$ has a minimum corresponding to some critical value of the parameter A. Physically this minimum corresponds to the critical pressure, below which the flow is blocked after the magnetic field application. Actually, if the applied pressure is less than the minimal value, the flow rate will decrease till zero after the magnetic field application, indicating the flow blockage. This is illustrated by the left vertical arrow in Fig. 7. So, the point S of the steady-state discharge curves at zero velocity ($A = 0$, Fig. 7) corresponds to the flow onset, when we have to apply some critical pressure to remove the plug from the capillary.

We see that the model predicts a hysteretic behavior of the discharge curve well verified in our experiments. First, the pressure required to stop the flow is less than the pressure required to restart the flow. Second, the flow onset and blockage are accompanied by velocity jumps denoted by two right vertical arrows in Fig. 7. Therefore within the pressure range between these two critical values, the suspension can either flow or be at rest. The reason of this hysteresis is the following: the applied pressure is used both to overcome the hydraulic resistance of the capillary and to push non-magnetic silica through the magnetic field, i.e. to overcome Stokes force of friction between silica and ferrofluid. If we increase the velocity from zero (point S), the hydraulic resistance increases, but the friction force decreases because the concentration profile becomes more uniform. The sum of both effects gives us the minimum of the pressure (blockage pressure) at some non-zero velocity, which is reached by decreasing the applied pressure.

It is important to note that, if we control the pressure, only the upper branch of the curve $p(A)$ is reachable. On the contrary, if we control the velocity and measure the pressure, the whole curve can be described and the starting point S at $A = v = 0$ corresponds to the static case where the pressure is applied after the formation of the plug. In this latter case we have a silica plug on the left of the magnets' axis $x = 0$ and the silica free zone on the right of this axis, the concentration profile being $\Phi_a = 0.64$ at $-1 < x < 0$ and $\Phi_a = 0$ at $0 < x < 1$. Therefore Eq. (28) will reduce to

$$p = -(1 - \Phi_a\phi)\int_{-1}^{0} g'(x)\,dx - 1 \cdot \int_{0}^{1} g'(x)\,dx = -(1 - \Phi_a\phi)\cdot 1 - 1\cdot(-1) = \Phi_a\phi.$$

Thus, we obtain the normalized pressure of the flow onset $p = \Phi_a \cdot \phi = \Phi_s^{pack} = 0.64 \cdot 0.64 \approx 0.41$, or in dimension form $\Delta P = \Phi_s^{pack}\mu_0 M_f^{sat} H_{max}$ with $\Phi_s^{pack} = \Phi_a \cdot \phi$ being the volume fraction of silica in the plug formed by aggregates of internal volume fraction ϕ. This result does not depend on the volume fraction Φ_0 of silica in suspension that is supported by our experiments. It is also consistent with our critical pressure estimation (cf. Eq. (10)) in the case when the plug is quite long and its rear border is subject to zero magnetic field ($H_1 = 0$ in Eq. (10)).

At high velocities the pressure tends to the one in the absence of the magnetic field (cf. dashed curves in Fig. 7). Theoretically the critical pressure in the case of the flow deceleration depends slightly on the initial concentration Φ_0, that intervenes into Eq. (27) (cf. solid curves in Fig. 7), but in practice we were not able to observe this difference in our experiments.

Finally we note that the existence of two equilibrium velocities for a given applied pressure is the indication of a structural instability that is often observed in the rheology of complex fluids, in particular those presenting a yield stress, which show a plateau in the shear stress versus shear rate curve (Volkova et al. (1999)). The fact that the change of structure with the flow can be described as a change in the concentration profile of aggregates and that the magnetic force acting on the

particles is known, has allowed us to do some quantitative prediction which represents fairly well the experimental behavior.

5. Conclusions

From this study we arrive at the following conclusions:

(1) The equilibrium position of a magnetic drop in a capillary placed between magnets is determined by the balance of external pressure and magnetostatic pressure in a suspension; it is shown that the critical pressure—when the drop is expelled from the high field domain—almost does not depend on the concentration of non-magnetic particles and is practically equal to the one of pure ferrofluid. For the saturation magnetization of 30 kA/m and a maximum induction of 0.75 T the critical pressure is 15 kPa.
(2) If, instead of a drop placed between the magnets, the whole capillary is filled with a ferrofluid containing non-magnetic particles, then two plugs form on each side of the high field region; when the pressure is increased these plugs move slowly till the back one passes the maximum of the field and is expelled suddenly. The critical pressure corresponding to this situation is lower than in the preceding case (about 10 kPa). This is because we have an inverse situation where it is the non-magnetic part, which creates the force in the inhomogeneous magnetic field. This force is thus proportional to the volume fraction of non-magnetic material inside the plug and the critical pressure is lowered in the same proportion; this is also the reason why the critical pressure is independent of the concentration of particles in a suspension (except for near zero concentration).
(3) When the field is applied in the presence of a flow—the case that would apply for blood embolism—the critical pressure for flow blockage is still less important that in the preceding case (about 5.5 kPa). For supercritical pressures, the flow rate decreases to some steady value. Decrease of the flow rate is mainly due to the friction between aggregates of particles and the ferrofluid and, for a smaller part to the rheological behavior of a suspension in a strong magnetic field. We have developed a model, which explains the main features of the experiments. In particular we predict a velocity versus pressure dependence presenting a hysteresis loop that explains the difference of behavior between critical pressures of the flow onset and the flow blockage.

Acknowledgments

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References

Bashtovoi, V.G., Mihalev, V.P., Reks, A.G., 1987a. Investigation of the behavior of a magnetic fluid in displacement and force transducers. Magnetohydrodynamics 23, 299–303.

Bashtovoi, V.G., Mihalev, V.P., Reks, A.G., Taitz, Ye.M., 1987b. Instability of finite volumes of magnetic fluid in channels. Magnetohydrodynamics 23, 49–53.

Bashtovoy, V.G., Berkovsky, B.M., Vislovich, A.N., 1988. Introduction to Thermomechanics of Magnetic Fluids. Hemisphere Publishing, New York.

Berthier, S., 1993. Optique des milieux composites. Polytechnica, Paris (in French).

Blums, E.Ya., Mayorov, M.M., Cebers, A.O., 1989. Magnetic Fluids. Zinatne, Riga (in Russian).

Bozorth, R.M., 1951. Ferromagnetism. van Nostrand, New York.

Drew, D.A., 1983. Mathematical modeling of two-phase flow. Ann. Rev. Fluid Mech. 15, 261–291.

Flores, G.A., Liu, J., 2001. Embolization of blood vessels as a cancer therapy using magnetorheological fluids. In: Bossis, G. (Ed.), Proceedings of the 8th International Conference on ER fluids and MR suspensions, Nice 9–13 July. World Scientific, pp. 146–152.

Jordan, A., Scholz, R., Maier-Hauff, K., Johannsen, M., Wust, P., Nadobny, J., Schirra, H., Schmidt, H., Deger, S., Loening, S., Lanksch, W., Felix, R., 2001. Presentation of a new magnetic field therapy system for the treatment of human solid tumors with magnetic fluid hyperthermia. J. Magn. Magn. Mater. 225, 118–126.

Kashevskii, B.Eh., Kordonskii, V.I., Prokhorov, I.V., 1988. Magnetorheological effect for nonmagnetic particles suspended in a magnetic liquid. Magnetohydrodynamics 24, 368–372.

Khalafalla, S.E., Reimers, G.W., 1973. Magnetogravimetric separation of nonmagnetic solids. AIME Trans. Soc. Mining Eng. 254, 193–198.

Khizhenkov, P.K., Bilorobov, V.M., Eremenko, S.V., 1993. Test of the application of magnetizable fluids and suspensions in experimental medicine. 2. Embolization of blood vessels. Magnetohydrodynamics 29, 97–98.

Lukashevich, M.V., Naletova, V.A., Tsurikov, S.N., 1988. Redistribution of the concentration of a magnetic fluid in a nonuniform magnetic field. Magnetohydrodynamics 24, 318–323.

Maruno, S., Yabakami, K., Soga, M., 1983. Plain paper recording process using magnetic fluids "Magneto-fluid-graphy". J. Magn. Magn. Mater 39, 187–191.

Rosensweig, R., 1985. Ferrohydrodynamics. Cambridge University Press, Cambridge.

Šafařík, I., Šafaříková, M., 1999. Use of magnetic techniques for the isolation of cells. J. Chromatogr. B 722, 33–53.

Taketomi, S., 1980. Motion of ferrite particles under a high gradient magnetic field in a magnetic fluid shaft seal. Jpn. J. Appl. Phys. 19, 1929–1936.

Taktarov, N.G., 1980. Motion of magnetizable liquids in porous media. Magnetohydrodynamics 16, 251–254.

Volkova, O., Cutillas, S., Bossis, G., 1999. Shear banded flows and nematic-to-isotropic transition in ER and MR fluids. Phys. Rev. Lett. 82, 233–236.

Volkova, O., Bossis, G., Guyot, M., Bashtovoi, V., Reks, A., 2000. Magnetorheology of magnetic holes compared to magnetic particles. J. Rheol. 44, 91–104.

Voltairas, P.A., Fotiadis, D.I., Michalis, L.K., 2002. Hydrodynamics of magnetic fluid targeting. J. Biomech. 35, 813–821.

Zick, A.A., Homsy, G.M., 1982. Stokes flow through periodic arrays of spheres. J. Fluid Mech. 115, 13–26.