Numerical modeling of ferrofluid flow instabilities in a capillary tube at the vicinity of a magnet

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1. ABSTRACT

The use of ferrofluids is now foreseen to be important for microfluidic systems and biotechnological microsystems as microvalves, micropumps or for the control and synchronization of the flow.

The advantage lies in the possibility of monitoring and controlling the flow inside the biochip by micro-magnets or micro-electromagnets located outside the microfluidic device.

A model for plug flow of ferrofluids separating reagents is presented here and an analysis of the instabilities of the flow is deduced from the model.

A comparison with experimental results is shown.

2. INTRODUCTION

Ferrofluids or magnetic liquids have been first developped in the 1960s as bearing seals for space applications. Later as sound damping systems; very recently, their utility for microfluidics systems has been demonstrated. For example, the principle of a magnetic pipette has been established in 1997 by Greivell and Hannaford [1] and that of a ferrofluidic microvalve in 1999 by Perez-Castillejos et al. [2].

New applications are foreseen in the domain of lab-on-a-chip and microarrays developped for biotechnological purposes.

In order to contribute to the development of such applications, we show here that caution should be used when dealing with ferrofluids in microsystems, specially when magnets and/or current loops are used to control the ferrofluid flow.

We examine the case of reagents flowing inside a cylindrical tube separated by one or more ferrofluidic liquid plugs (fig 1 and fig 2). The flow is established by a driving pressure at the inlet and its velocity is controled by one or more magnets. It will be shown by a numerical model that instabilities can appear in the vicinity of the magnet, the result being unexpected break-ups of the ferrofluid plug into smaller plugs. Experiments have been performed that confirm the numerical simulations. Such an event is a serious drawback for biotechnological applications because it results in the mixing of the liquid reagent between different plugs.

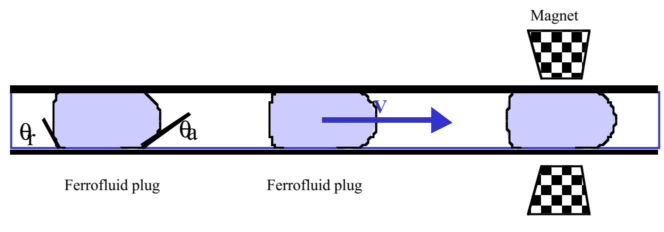

Fig 1. Flow of ferrofluid plugs and reagent passing by a magnet. θ_a, resp. θ_r are the advancing and receeding angles.

Fig. 2. Photograph of ferrofluid plugs separating reagents flowing in a capillary tube.

3. NUMERICAL MODEL

As the flow circulates in a capillary tube, it is legitimate to make use of Bernoulli's equation. The numerical model is thus based on the Bernoulli's equation for ferrofluids that was established first by Rosenszweig [3]. The total pressure drop in the channel is the sum of the capillary, friction and magnetic pressure drop

$$\Delta P_{channel} = \Delta P_{capillary} + \Delta P_{friction} + \Delta P_{magnetic} \qquad (1)$$

3.1. Friction pressure drop

The model uses the classical Washburn law [4] for the pressure drop due to friction at the solid walls.

$$\Delta P_{friction} = \frac{8V}{\pi R^2}\left(\eta_1 L_1 + \eta_2 L_2\right) \qquad (2)$$

where indices 1 and 2 points to liquid 1 (ferrofluid) and liquid 2 (reagent). R is the radius of the capillary, V the liquid velocity and L_1,L_2 the total length of liquid 1,2 plugs.

3.2. Capillary pressure drop

The capillary pressure drop is due to the difference of the capillary forces between advancing and receeding fronts because of the two different contact angles (advancing and receeding) θ_a et θ_r

$$\Delta P_a = \sum_{plugs} \frac{2\sigma}{R} \cos\theta_a \quad (3)$$

$$\Delta P_r = \sum_{plugs} \frac{2\sigma}{R} \cos\theta_r \quad (4)$$

The total capillary pressure drop is then

$$\Delta P_{capillary} = \frac{2\sigma}{R} N_{plugs} \left(\cos\theta_a + \cos\theta_r \right) \quad (5)$$

From (5) it can be seen that too many plugs in the capillary will block the flow. In the model, the capillary pressure drop is based on the Hoffman-Tanner law [5]

$$\begin{aligned} \theta_a^3 &= \theta_{a,s}^3 - \beta\, Ca_1 \\ \theta_r^3 &= \theta_{r,s}^3 - \beta\, Ca_2 \end{aligned} \quad (6)$$

where the index s stands for the static contact angle, β is a constant (of the order of 60) and Ca_1, Ca_2 are respectively the capillary numbers for liquid 1 and 2

$$\begin{aligned} Ca_1 &= \frac{V\eta_1}{\sigma} \\ Ca_2 &= \frac{V\eta_2}{\sigma} \end{aligned} \quad (7)$$

Eq (6) assumes that the static contact angle are uniquely defined , when this is not the case they will be defined as the limit of the dynamic contact angles with vanishing velocity. Hoffman-Tanner law states that contact angles decrease with increasing velocity. Because the capillary number is small in most biochips applications – a typical value is of the order of 10^{-3} -, the Hoffman-Tanner law can be linearized using a Taylor expansion of order 1.

$$\begin{aligned} \theta_a &= \left(\theta_{a,s}^3 - \beta\, Ca_1 \right)^{\frac{1}{3}} \cong \theta_{a,s} \left(1 - \frac{1}{3} \frac{\beta\, Ca_1}{\theta_{a,s}^3} \right) \\ \theta_r &= \left(\theta_{r,s}^3 - \beta\, Ca_1 \right)^{\frac{1}{3}} \cong \theta_{r,s} \left(1 - \frac{1}{3} \frac{\beta\, Ca_2}{\theta_{r,s}^3} \right) \end{aligned} \quad (8)$$

and the cosine of the advancing and receeding angle can advantage that it permits to obtain an expression of the capillary pressure drop directly as a function of the velocity

$$\Delta P_{capillary} = \frac{2\sigma}{R} N_{plugs} \left(\cos\theta_{a,s} + \cos\theta_{r,s} \right) + \frac{2\beta V}{3R} N_{plugs} \left(\frac{\eta_1}{\theta_{a,s}^2} \sin\theta_{a,s} + \frac{\eta_2}{\theta_{r,s}^2} \sin\theta_{r,s} \right) \quad (9)$$

The first term of eq. (9) depends on the difference between the static contact angles, the second term is proportionnal to the velocity.

3.3. Magnetic pressure drop

The magnetic forces exerted on the ferrofluidic plug are given by Rosensweig's law taking into account the magnetic pressure as well as the normal magnetic pressure. For one ferrofluid plug, one obtains

$$\Delta P_{magnetic} = \mu_0 \int_{H_r}^{H_a} M\, dH + \frac{\mu_0}{2} \left(M_{n,a}^2 - M_{n,r}^2 \right) \quad (10)$$

where H is the magnetic field along the capillary axis, M the magnetic moment and M_n is the magnetic moment normal to the plug interface with water.

Now if we remark that a ferrofluid behaves like a paramagnetic media [6], the Langevin's law applies [7]

$$\frac{M}{M_s} = \coth\left(\frac{3\chi H}{M_s}\right) - \frac{1}{\frac{3\chi H}{M_s}} \quad (11)$$

where M_s is the saturation magnetization and χ the magnetic susceptibility of the ferrofluid. In the limit of low magnetization $M = \chi H$ and one obtains from (10), for one plug

$$\Delta P_{magnetic} = \frac{1}{2} \frac{\mu}{\mu_0} \left(\mu - \mu_0 \right) \left(H_a^2 - H_r^2 \right) \quad (12)$$

This formula shows that it is the square of the magnetic field at the interfaces that defines the magnetic force.

For the full Langevin's formulation (11), after integration of the first term of the left hand side of eq. (10), one obtains

$$\Delta P_{magnetic} = \frac{\mu_0 M_s^2}{3\chi}\left[Ln\left(\frac{sh\left(\frac{3\chi H}{M_s}\right)}{\frac{3\chi H}{M_s}} \right)\right]_{Hr}^{Ha} + \frac{\mu_0 M_s^2}{2}\left[\left(\frac{coth\left(\frac{3\chi H}{M_s}\right)}{\frac{3\chi H}{M_s}} \right)^2\right]_{Hr}^{Ha} \qquad (13)$$

It will be shown that it is not necessary to make the summation of the magnetic pressure drop over all the plugs because of the very sharp decrease of the magnetic field with the distance.

4. ANALYSIS OF THE NUMERICAL RESULTS

The model has been developped under Matlab. Given a driving pressure drop, the velocity is calculated using eq. (1), (2), (9) and (12) or (13). Location of the plug inyterfaces should be carefully calculated in order to obtain an accurate result.

The model shows sharp velocity changes in the vicinity of the magnet (fig. 2 and 3) due to sudden accelerations and decelerations of the flow.

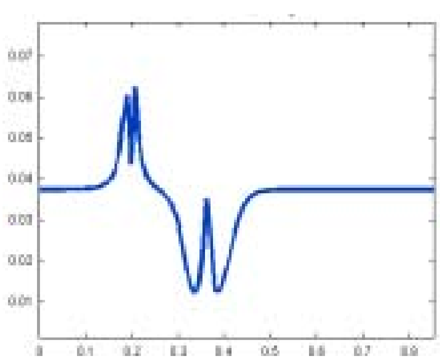

Fig 2. Velocity of the flow when a ferrofluid plug passes near the magnet. In this case the plug length is large compared to the extent of the magnetic field

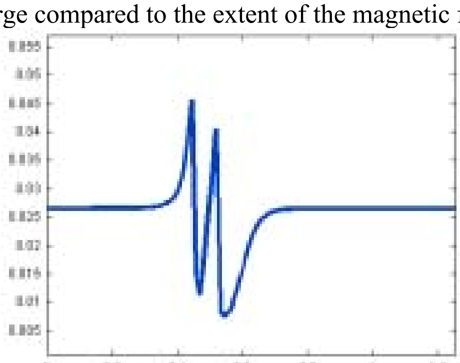

Fig 3. Velocity of the flow when a ferrofluid plug passes near the magnet. In this case the plug length is small compared to the extent of the magnetic field

The plug is at first accelerated when approaching the magnet - this is clearly seen on experiments (fig. 5). The different velocity peaks (4 for a plug of a sufficiently long extent, 3 for a short plug) can be explained physically. For a "long" plug, i.e. a plug of a longer extent than that of the magnetic field, only the advancing front of the plug is first concerned by the magnetic force, and the velocity varies accordingly to the field H^2 (as can be seen from eq. 12), then only the receeding front is affected by the magnetic force and the velocity is related to the field $-H^2$. The profile of the square of the magnetic field is shown in fig 4. The similarity with the velocity profile is clear.

For a "short" plug the same reasonning applies except that one has to take into account the advancing and the receeding fronts. In fact, the preceeding explanation yields for the first and fourth peaks of velocity, but the second and third peaks collapse to a unique velocity oscillation due to the joint effect of the magnetic forces on the two interfaces.

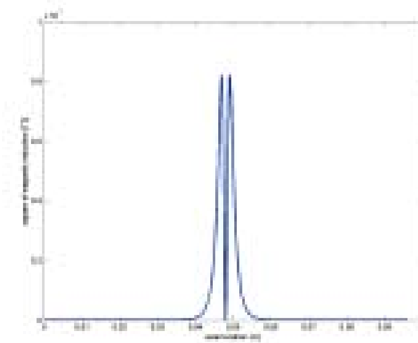

Fig. 4. Profile of the square of the magnetic field of the magnet.

In the case where one wants to stop completely the flow, the magnet has to be placed sufficiently close to the channel in order to deliver a magnetic field large enough. In such a case, the magnetic linear model does not apply and and the full Langevin's model is relevant. For example, in fig. 5, the model shows that the flow is not stopped by the magnet contrary to the result of the simplified linear model.

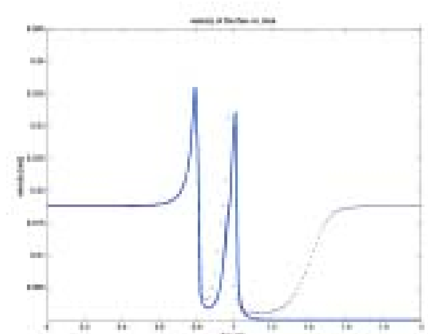

Fig5. Velocity vs. time for a "short" plug. The flow is predicted to be stopped by the simplified model but not by the full magnetic model (dot line)

5. EXPERIMENTS

Experiments have been performed using teflon or glass cylindrical capillary tubes of 500 and 320 µm of inner diameter. Two types of ferrofluids have been used: one with an organic base (EMG901 from Ferrofluidics Corp.) and another one with a ionic base (from the "Laboratoire des Liquides Ioniques et Interfaces Chargées" de l'Université Paris 6). Ionic base ferrofluids are performing better because organic base ferrofluids are hydrophobs and tend to leave a film on teflon coating. A permanent magnet (neodym-iron-bore) of 0.5 x 5 x 36 mm is placed perpendicularly to the capillary.

The driving pressure is of the order of 1000 to 3000 Pa, which is correctly predicted by the model. It depends on the number of plugs and the velocity of the fluid.

Fig 6 shows the nearly break up of a ferrofluid plug when its advancing front approaches the magnet. This is related to the velocity increase of the flow at the approach of the magnet predicted by the theoretical model.

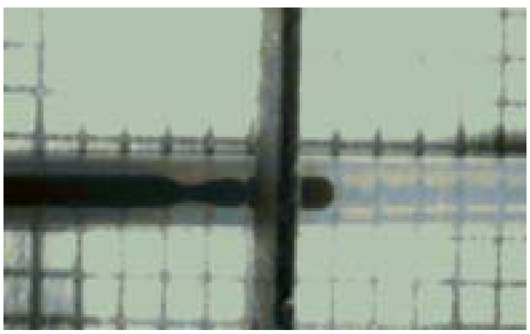

Fig. 6. The acceleration of the advancing front of the ferrofluid plug (in dark) at the approach of the magnet provoques a near break up of the plug.

In the case where the magnetic force is not sufficient to block the flow, the ferrofluid plug can be dispersed into annulae with reagent flowing in the middle (fig. 7). This is related to the sudden velocity decrease of the plug after it has passed the magnet accordingly to the prediction of the theoretical model.

It is therefore important to optimize the magnetic field used to control the flow. It can be shown that a solenoid performs better than a permanent micro-magnet.

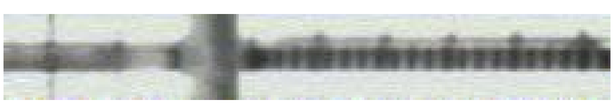

Fig. 7. Break up of the ferrofluid plug due to the decceleration of the receeding front at the magnet location. The reagent flows in the middle of the capillary through the annulus of ferrofluid.

6. CONCLUSION

Instabilities of ferrofluid plugs inside capillaries are due to sudden changes in the velocity of the flow when a plug passes near the magnet. These instabilities, observed by experiments in 320 and 500 µm section capillary tubes, are predicted by the numerical model.

These instabilities appear when the flow is slowed ferrofluid plug, but not totally stopped, or when the flow is stopped and the magnet is withdrawn in order to release the flow in the capillary.

The magnetic force on a ferrofluid plug is proportional to the square of the magnetic field. Thus it is of great importance to optimize the magnetic field in order to reduce the spatial variation of the magnetic force.

7. REFERENCES

[1] N.E. Greivell; B. Hannaford . "The design of a ferrofluid magnetic pipette". IEEE-Transactions-on-Biomedical-Engineering. vol.44, no.3; March 1997; p.129-35

[2] R. Perez-Castillejos, J.A. Plaza, J . Esteve, P. Losantos, M.C. Acero, C. Cané, F. Serra-Mestres. "The use of ferrofluids in micromechanics". Sensors and Actuators, 84, pp 176-180, 2000.

[3] R.E. Rosensweig. "Ferrohydrodynamics". Cambridge University Press, New-York, 1985

[4] E.W. Washburn. "The dynamics of capillary flow". Phys. Rev., :273-283, 1921.

[5] M. Fermigier, P. Jenffer. "An experimental investigation of the dynamic contact angle in liquid-liquid systems". Journal of Colloid and Interface Science, vol 146, pp226-241, 1990.

[6] A. Skumiel, T. Hornowski, A. Jozefczak. "Acoustic and magnetic properties of magnetic liquids". Archives of Acoustics, 24, n°2, pp 129-144, 1999.

[7] R.W. Chantrell, J. Popplewell, S.W. Charles, "Measurements of particle size distribution parameters in ferrofluids". IEEE Trans. On Mag. Vol MAG-14, n°5, pp 975-977, 1978.