

# Optimization of magnetic actuation protocol to enhance mass transfer in solid/liquid microfluidic systems

Evgeny REBROV<sup>1,2</sup>, Erell BONNOT<sup>1</sup>, Volkan DEGIRMENCI<sup>1</sup>, Robert POLLARD<sup>3</sup>, Robert BOWMAN<sup>3</sup>

\* Corresponding author: Tel.: ++44 (0)24 765 22202; Email: [e.rebrov@warwick.ac.uk](mailto:e.rebrov@warwick.ac.uk)

<sup>1</sup> School of Chemistry and Chemical Engineering, Queen's University Belfast, Belfast, UK

<sup>2</sup> School of Engineering, University of Warwick, Coventry, UK

<sup>3</sup> School of Mathematics and Physics, Queen's University Belfast, Belfast, UK

## Abstract:

The dynamic properties of a 250  $\mu\text{m}$  magnetic microparticle in a time varying magnetic field have been studied in a PDMS microreactor with a diameter of 13 mm using a dual coupled quadrupolar arrangement of electromagnets. A sinusoidal applied magnetic field has dictated a circular motion of the particles in the microreactor in the frequency range below 0.6 Hz. Different circular motion modes have been observed at higher frequencies of the applied field. The particular symmetric arrangement of the magnets has allowed a non-steady-state motion with variation in velocity between magnetic poles. The motion of magnetic particle has been described in terms of average velocity and mean square deviation from average velocity. The effect of actuation protocol parameters (frequency, magnetic field strength and phase shift) on particle velocity and acceleration has been investigated. The maximum average velocity of 0.016 m/s has been observed under an optimized actuation protocol. The mass transfer rate towards the particle surface is mainly influenced by the average velocity while the effect of acceleration/deceleration of the particle has an order of magnitude less influence.

**Keywords:** Magnetic microparticles, magnetic actuation, quadrupolar, microreactor.

## 1. Introduction

Recently in the expanding field of microfluidics the drive to lab on a chip systems has promised more efficient chemical processes on a smaller scale. Magnetic micro/nano particles with the promise of a remote method of manipulation and larger surface areas have opened up a wide number of potential applications in this field<sup>1-3</sup>. Catalysis using magnetic microparticles has proved to be one of the most exciting possibilities and is growing into one of the most promising applications within the field of nanoscience. The large relative surface area of microparticles qualifies them quite naturally to act as a support for homogeneous and heterogeneous catalysts. Contrary to classic heterogeneous catalysts, catalytic nanoparticles are synthesized in a bottom-up approach from molecular precursors such as a metal salt, and a reducing agent. In the field of biomedicine these microparticles have also been proposed as a source of both drug delivery and targeting as an

alternative to conventional chemotherapy drugs<sup>4,5</sup>, and as a hyperthermia agent<sup>6</sup>. Specific engineering of the particles surface has also allowed for capture and binding of targeted species<sup>7</sup>.

Several magnetic actuation systems have been demonstrated such as a rotating system of magnets allowing for magnetophoretic separation of magnetic particles<sup>8</sup> and a pair of magnetic tweezers for active control of magnetic particles<sup>9</sup>. Magnetophoresis of nanoparticles has also produced promising results using dark field microscopy<sup>10</sup>. A quadrupolar arrangement of electromagnets is often used by researchers for magnetic actuation of microparticles. For example, by combining alternating and static magnetic fields (two permanent magnets and an electromagnet), cyclic motion of superparamagnetic beads has been induced<sup>11</sup>. Rotational dynamics of magnetic particles has been obtained by applying sinusoidal electric signals to two pairs of coils. Feedback control has also been demonstrated which is based

on video monitoring the magnetic droplet location and then providing a correcting action by adjustments to the actuation protocol<sup>12</sup>.

If the particles are completely immersed in liquid, an increase in the stirrer speed leads to a thinning of the laminar liquid film that adheres to the particles. Therefore, the liquid side mass transfer rate can be improved. Mass transfer is usually described by the Sherwood number ( $Sh$ ) which is a function of the Reynolds number ( $Re$ ) and the Schmidt number ( $Sc$ ).

$$Sh = 2 + 0.6Re_p^{1/2}Sc^{1/3} \quad \text{for } Re_p > 1 \quad (1)$$

$$Re_p = \frac{vd_p}{\nu} \quad (2)$$

$$Sc = \frac{\nu}{D} \quad (3)$$

where  $v$  is the average velocity of the microparticle in the surrounding fluid,  $\nu$  is the kinematic viscosity of the fluid,  $d_p$  is the particle diameter, and  $D$  is the diffusion coefficient of reactant. The  $Sc$  number equals to 243 for diffusion of a  $C_5$  organic alcohol in acetonitrile, considered here as a model reaction. To intensify mass transfer it is necessary to influence the hydrodynamics around the microparticle though controlling the relative velocity. An increase in relative velocity leads to a reduction of the laminar boundary layer thickness around the particle, and thus, to an increase in the mass transfer coefficient.

The goal of this study is to optimise the actuation protocol for manipulation of magnetic particles to achieve maximum enhancement of reactant mass transfer towards the surface of a catalytic particle in a chemical reaction. To reach this goal particle movement induced by the various cycles of rotating magnetic field was studied and subsequently a design strategy was proposed to efficiently manipulate microparticle in a flow field at  $Re_p > 1$ .

## 2. Experimental Method

The magnetic actuation setup consists of a quadrupolar set of iron bars, 14 cm long with a cross section of  $2.5 \times 2.5$  cm<sup>2</sup> connected to four horizontal coils. The coils, connected to 2 separate direct current (DC) power supplies (Kepco BOP 100-2ML) were coupled in two perpendicular pairs by two conductive iron bases. Sinusoidal voltages

( $V_1, V_2$ ) were applied to the A and B pairs of the electromagnets, respectively, using a Labview software:

$$V_1 = V_{01} \cdot \sin(\omega t) \quad (4)$$

$$V_2 = V_{02} \cdot \sin(\omega t + \phi) \quad (5)$$

where  $V_{01}$  and  $V_{02}$  are the maximum applied voltages through the both pairs of magnets,  $\omega$  is the frequency of the time dependent magnetic field and  $\phi$  is the phase shift. A 13 mm diameter PDMS reactor filled with acetonitrile was placed over the X-Y stage of an optical microscope (Leica M165 FC) connected to a video camera (Leica DFC310 FX) with a resolved pixel size of  $6.5 \times 6.5$   $\mu\text{m}^2$ . A single microparticle of 250  $\mu\text{m}$  in diameter was placed in the center of the reactor. The actuation protocols were developed in LabView to control the voltage applied to the coils, and so the magnetic field. The maximum magnetic field strength was 145 kA m<sup>-1</sup>. The image analysis was performed using a NI-Vision software. Images were taken at a rate of 20 fps allowing observation of the particle motion in an area of 1060 mm<sup>2</sup>. For each set of parameters ( $V, \omega, \phi$ ), the particle trajectory of was recorded over several periods of rotation and the local velocity, local acceleration and the average standard deviation of the velocity were calculated.

## 3. Results and discussion

Due to a non-uniformity of magnetic gradient, the particle experiences four acceleration and deceleration phases during a single period of rotation in the reactor. To optimize the actuation protocol, the following objective function was introduced:

$$f = a_1 \tilde{v} + a_2(1 - \tilde{\sigma}) \quad (6)$$

where  $\tilde{v}$  is the dimensionless average velocity which is the ratio of the average velocity over 10 rotational cycles to the maximum velocity of 1.6 cm s<sup>-1</sup> observed in this study (Eq. 7);  $\tilde{\sigma}$  is the dimensionless standard deviation of the velocity (Eq. 8) which is the ratio of the standard deviation corresponding to a given trajectory (Eq. 9) to the maximum standard deviation observed in this study ( $\sigma_{v,\max}$ ).

$$\tilde{v} = \frac{\bar{v}}{\bar{v}_{\max}} \quad (7)$$

$$\tilde{\sigma} = \frac{\sigma_v}{(\sigma_v)_{\max}} \quad (8)$$

$$\sigma_v = \sqrt{\frac{1}{n-1} \sum_{i=1}^n (v_i - \bar{v})^2} \quad (9)$$

As both the absolute velocity and the acceleration influence the boundary layer thickness and therefore the mass transfer rate, their contributions to objective function  $f$  were taken into account via weight factors  $a_1$  and  $a_2$ , respectively.

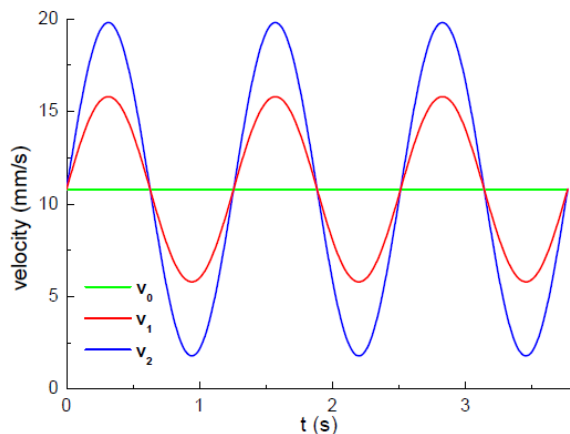
$$a_1 = \frac{R_1}{R_1 + R_2}, \quad (10)$$

$$a_2 = \frac{R_2}{R_1 + R_2}, \quad (11)$$

where index  $R_1$  is defined as the difference between the maximum and minimum Sh numbers observed in this study (Eq. 12) and index  $R_2$  is defined as the difference between the time averaged Sh numbers corresponding to non-steady motions of a magnetic microparticle with maximum and minimum standard deviations from the average velocity as shown in Figure 1 (Eq. 13).

$$R_1 = Sh_{\max} - Sh_{\min}, \quad (12)$$

$$R_2 = \frac{1}{t_1} \int_0^{t_1} (Sh_{\sigma_{\max}}(t) - Sh_{\sigma_{\min}}(t)) dt \quad (13)$$

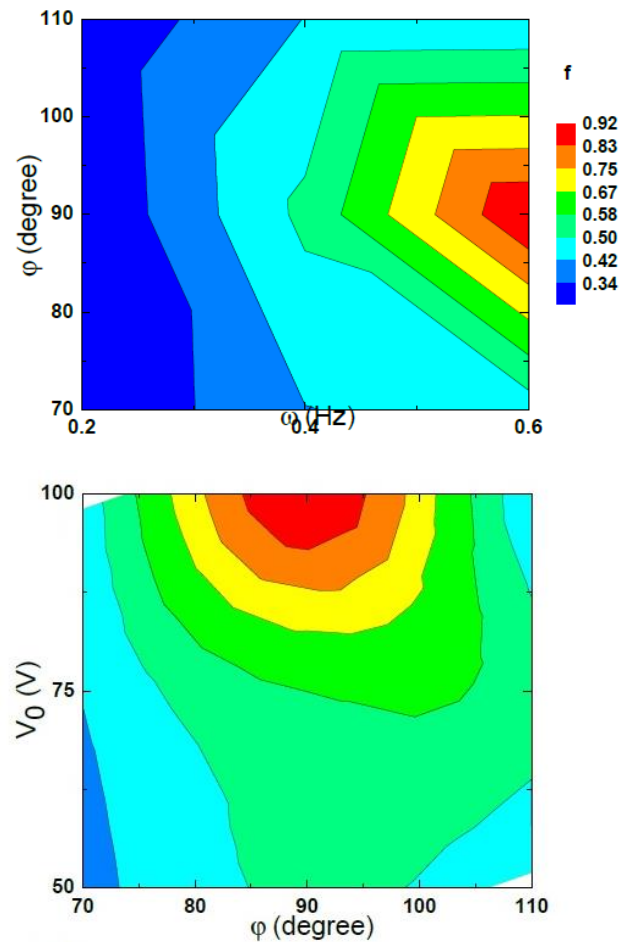


**Figure 1:** Particle velocity as a function of time for three cases corresponding to the maximum ( $v_2$ ) and minimum ( $v_1$ ) standard deviation from the average value, and a steady state velocity profile ( $v_0$ ).

Any deviations from the average velocity would decrease Sh number, the larger  $\tilde{\sigma}$ , the smaller would be mass transfer rate. To calculate  $R_2$ , the functional dependencies of particle velocity,

corresponding to the maximum and minimum standard deviation (Fig. 1) were inserted in Eq. 1 and the corresponding time averaged Sherwood numbers,  $Sh_{\sigma_{\max}}(t)$  and  $Sh_{\sigma_{\min}}(t)$ , were calculated over a single period of particle rotation. The obtained values of  $R_1$  and  $R_2$  are equal to 7 and 0.5, respectively. This means that the effect of average velocity on Sh number exceeds that of non-steady motion by an order of magnitude. It can also be seen from the normalised weight factor values and of 0.93 and 0.07 for  $a_1$  and  $a_2$  respectively.

A parametric study was performed to find the maximum of objective function  $f$ . The design parameters were changed as follows: the frequency was changed between 0.2 and 0.6 Hz, the maximum voltage on the coils was changed in the range from 50 to 100 V and the phase shift between the two sets of coils was varied from 70 to 110°.



**Figure 2:** Objective function  $f$  as a function of frequency and phase shift at a maximum voltage on the coils of 100 V (top) and phase shift voltage and at a frequency of 0.6 Hz (bottom).

The maximum of function  $f$  was found at a

frequency of 0.6 Hz, a voltage of 100 V and a phase shift of 90° (Figure 2). Higher frequencies change the dynamics of particle motion. While the local velocity can still be increased even further, the particle does not follow the circular trajectory near the perimeter of the cell, but it starts non-periodic oscillations near a single magnetic pole. Therefore operating frequencies above 0.6 Hz were not considered in this study. There seems to be a fast response of the particle motion to the applied magnetic field in acetonitrile as the optimized value of phase shift is 90°. That means there is no need for any delay in phase shift to account for inertia of the magnetic particle.

The sensitivity analysis shows that objective function  $f$  remains within 5% from its maximum value if the maximum voltage on the coils drops to 95 V (or by 5% from the optimum value). The effect of phase shift has a similar impact: as the phase shift increases (or decreases) by 4° from the optimum value, the objective function value reduces from 0.94 to 0.88. A variation in magnetic susceptibility of the particle within 10% does not shift the optimum values of design parameters.

(a)

(b)

## 4. Conclusions

A motion of a 250 μm magnetic microparticle in a circular microfluidic cell with a diameter of 13 mm placed in a quadrupolar magnetic actuation setup has been studied. As the particle velocity increases, the motion pattern changes from a circular motion to an oscillation near a single magnetic pole pattern. The circular motion was observed in the range of average particle velocities between 0.2 and 1.6 cm/s. During a single period of rotation in the cell, the particle experiences a time dependent magnetic field gradient and its motion has four acceleration and deceleration cycles. Therefore the particle motion has been described in terms of the average velocity and the standard deviation from the average velocity. The both motion parameters depend on magnetic actuation protocol parameters such as the frequency of magnetic field, phase shift between the two sets of coils and the intensity of magnetic field. To get the maximum mass transfer rate in a chemical reaction, the actuation protocol parameters corresponding to the maximum average velocity and the minimum standard deviation have been found. A frequency of 0.6 Hz, a phase shift of 90° and a voltage of 100 V provide the maximum mass transfer rate. Under these conditions the

effect of the average velocity magnitude on the enhancement of mass transfer rate was an order of magnitude larger than that of acceleration/deceleration during the motion.

**Acknowledgment:** The financial support provided by the European Research Council (ERC) project 279867, is gratefully acknowledged.

## References

1. Gijs, M.A.M. 2004. Magnetic bead handling on-chip: New opportunities for analytical applications *Microfluidics and Nanofluidics* **1**, 22-40
2. Gijs, M.A.M., Lacharme, F., Lehmann, U. 2010. Microfluidic applications of magnetic particles for biological analysis and catalysis *Chem. Reviews* **110**, 1518-1563.
3. Hsing, I.M., Xu, Y., Zhao, W. 2007. Micro- and nano- magnetic particles for applications in biosensing *Electroanalysis*, **19**, 755-768.
4. Nacev, A., Beni, C., Bruno, O., Shapiro, B. 2010. Magnetic nanoparticle transport within flowing blood and into surrounding tissue *Nanomedicine* **5**, 1459-1466.
5. Nacev, A., Beni, C., Bruno, O., Shapiro, B. 2011. The behaviors of ferromagnetic nano-particles in and around blood vessels under applied magnetic fields *J Magnet. Mater.* **323**, 651-668.
6. Goya, G.F., Lima, E.Jr., Arelaro, A.D., Torres, T.E., Rechenberg, H.R., Rossi, L., Marquina, C., Ibarra, M.R. 2008. Magnetic hyperthermia with Fe<sub>3</sub>O<sub>4</sub> nanoparticles: The influence of particle size on energy absorption. *IEEE Trans. on Magnetics* **44**, 4444-4447.
7. Veisheh, O., Gunn, J.W., Zhang, M. 2010. Design and fabrication of magnetic nanoparticles for targeted drug delivery and imaging. *Adv. Drug Delivery Rev.* **62**, 284-304.
8. Andreu, J.S., Camacho, J., Faraudo, J., Benelmekki M, Rebollo C, MartÁnez LM (2011) Simple analytical model for the magnetophoretic separation of superparamagnetic dispersions in a uniform magnetic gradient. *Physical Review E* **84**, 021402.
9. Zhang, Z., Huang, Y., Menq, C.H. 2010. Actively controlled manipulation of a magnetic microbead using quadrupole magnetic tweezers. *IEEE Trans. on Robotics* **26**, 531-541.
10. Lim, J., Lanni, C., Evarts, E.R., Lanni, F., Tilton, R.D., Majetich, S.A. 2011. Magnetophoresis of nanoparticles. *ACS Nano* **5**, 217-226.

11. Moser, Y., Lehnert, T., Gijs, M.A.M. 2009. Quadrupolar magnetic actuation of superparamagnetic particles for enhanced microfluidic perfusion. *Appl. Phys. Lett.* **94** 022505.
12. Probst, R., Lin, J., Komae, A., Nacev, A., Cummins, Z., Shapiro, B. 2011. Planar steering of a single ferrofluid drop by optimal minimum power dynamic feedback control of four electromagnets at a distance *J. Magnet. Magnet. Mater.* **323**, 885-896.