S1. VALIDITY OF BEER-LAMBERT'S LAW

The aqueous suspension of iron oxide magnetic nanoparticles (MNPs) coated with polyethylene glycol (PEG), which was used in the present investigation, was purchased from Ocean NanoTech. The received MNPs solution with initial concentration of 1000 mg/L, was diluted to 1 mg/L by deionized (DI) water obtained from Elga Purelab Flex with resistivity of 18.2 M Ω cm. Three mL of diluted MNPs solution was filled into a standard 1 × 1 × 4 cm disposable cuvette. UV-vis spectrophotometer (Agilent Cary-60) with monochromatic light beam at wavelength of 530 nm was employed to measure light absorbance of the MNPs solution (which was filled in the cuvette). The procedure above was repeated by diluting the received MNPs solution to other concentrations: 2, 3, 4, 5, 10, 20, 30, 50, 80 and 100 mg/L such that light absorbance of the MNPs solution with different concentration was recorded.

The graph of light absorbance versus MNPs concentration is displayed in Figure S1. It is observed that a straight line fit almost perfectly into the data points recorded in the experiment described above, with coefficient of determination $R^2 = 0.999$. Consequently, it can be deduced that the linear dependency of light absorbance on MNPs concentration is true for MNPs concentration which is ranging from 1 to 100 mg/L. This analysis indicates the validity of Beer-Lambert's Law as shown below:¹

$$A = \varepsilon lc \tag{S1}$$

where A is the light absorption of MNPs solution, ε is the light absorptivity of MNPs, l is the optical path length and c is the concentration of MNPs solution. Since our experiments are carried out within this concentration range, the linear dependency of light absorbance on MNPs concentration is assumed in the interpretation and analysis of the experimental result.



Figure S1. Plot of light absorbance versus MNPs solution concentration. The nearly perfect fitting of dotted straight line shows that light absorbance varies linearly with MNPs concentration.

S2. COMPARISON BETWEEN THE AMOUNT OF METHYLENE BLUE (MB) MOLECULES AND MAGNETIC NANOPARTICLES (MNPs) IN DYE EXPERIMENT

The highest MNPs concentration adopted in current study (100 mg/L) is assumed in the following calculation. According to the specification sheet provided by supplier, the molar MNPs concentration (in mol/L) of the received MNPs solution (which concentration is 1000 mg MNPs/L) is given by:

$$C_{MNPs.o} = 3.4 \times 10^{-8} mol/L$$

After diluting the received MNPs solution to 100 mg/L, the molar MNPs concentration (in mol/L) is reduced to:

$$C_{MNPs,100mg/L} = 3.4 \times 10^{-9} mol/L$$

Therefore, in 3 ml of 100 mg/L (by mass) MNPs solution, the amount (in term of number of mole) of MNPs is:

$$N_{MNPs} = C_{MNPs,100mg/L} \times V$$

= 3.4 × 10⁻⁹ $\frac{mol}{L}$ × 10⁻³ $\frac{L}{mL}$ × 3 ml
= 1.02 × 10⁻¹¹ mol

In order to track the fluid flow throughout magnetophoresis, approximately 0.05 mL of 3000 mg/L Methylene Blue (MB) solution was injected into the cuvette filled with 3 mL of MNPs solution. As the molecular weight of MB is 319.85 g/mol, the number of mole of MB molecules injected into the MNPs solution is calculated as:

$$N_{MB} = \frac{3000 \frac{mg}{L} \times 10^{-3} \frac{L}{mL} \times 10^{-3} \frac{g}{mg} \times 0.05 \, mL}{319.85 \frac{g}{mol}}$$
$$= 4.69 \times 10^{-7} \, mol$$

$$\frac{N_{MB}}{N_{MNPs}} = \frac{4.69 \times 10^{-7} \, mol}{1.02 \, \times 10^{-11} \, mol} \approx 45977$$

 N_{MB}

Since N_{MNPs} is far greater than unity, amount of MB molecules is overwhelming the amount of MNPs in the MNPs solution used in dye experiment ($N_{MB} >> N_{MNPs}$). Even though MB molecules and MNPs are carrying opposite charge and some MB molecules might attach to MNPs due to electrostatics attraction,² there is still a huge amount of excess MB molecules which are freely suspended in the solution and function as tracer to help us visualize fluid motion within the MNPs solution when it is subjected to an external magnetic field.

S3. MAGNETIC BJERRUM LENGTH AND AGGREGATION PARAMETER ANALYSIS

Magnetic Bjerrum length. Magnetic Bjerrum length is the distance between two parallel magnetic dipoles where magnetic energy and thermal energy are equal. It is defined as:³

$$\lambda_B = \left[\frac{8\pi\mu_0 M_{p,\nu}^2}{9k_B T}\right]^{1/3} R^2$$
(S2)

where λ_B is magnetic Bjerrum length, μ_0 is permeability of free space (= $4\pi \times 10^{-7}$ H m⁻¹), $M_{p,v}$ is volumetric magnetization of particle, k_B is Boltzmann's constant (= 1.381×10^{-23} J/K), T is absolute temperature and R is particle radius. For the MNPs system used in our study, magnetic Bjerrum length is calculated as follows:

 $M_{p,v} = 1.474 \times 10^5 A/m$ [Saturation magnetization is assumed]

- T = 300 K
- R = 15 nm

$$\lambda_B = \left[\frac{8\pi (4\pi \times 10^{-7})(1.474 \times 10^{5})^2}{9(1.381 \times 10^{-23})(300)}\right]^{\frac{1}{3}} (15 \times 10^{-9})^2 = 5.940 \times 10^{-8} m$$
$$= 59.4 \ nm$$

The calculation shows that the magnetic energy is equal to thermal energy when two fully magnetized MNPs are separated by a distance of 59.4 nm. However, the average interparticle spacing, d_p of MNPs used in the study is given by:³

$$d_p = \left(\frac{4\pi R^3 \rho_p}{3c}\right)$$
$$= \left(\frac{4\pi (15 \times 10^{-9})^3 (3455.43)}{3(100 \times 10^{-3})}\right)^{\frac{1}{3}}$$
$$= 7.876 \times 10^{-7} \, m = 787.6 \, nm$$

where ρ_p is the particle density. In this calculation, the highest MNPs concentration (which is c = 100 mg/L) employed in this experiment is assumed. According to the result of calculation, it can be shown that the average interparticle spacing of MNPs in the most concentrated MNPs solution used in current investigation is given by 787.6 nm and it is about 13.25 times greater than the magnetic Bjerrum length. Since the ratio λ_B/d_p is much greater than unity, MNPs/MNPs interaction and formation of reversible aggregates is not significant in the MNPs solution used in this experiment. Henceforth, MNPs behave independently with respect to each other throughout the magnetophoresis process.

Aggregation parameter. Aggregation parameter, N* is developed to describe the degree of interaction between suspended particles, which exhibit net magnetization, in a solution and it is formulated as follows:^{4, 5}

$$N^* = \sqrt{\phi_o e^{\Gamma - 1}} \tag{S3}$$

where ϕ_0 is the volume fraction of particles in the solution and Γ is magnetic coupling parameter. Magnetic coupling parameter is the parameter that characterizes the significance of magnetic interaction between two magnetic particles suspended in a solution and it is defined as the ratio of magnetic energy to thermal energy when the two particles are at the close contact:⁴

$$\Gamma = \frac{\mu_0 M_{p,\nu}^2 V_p}{12k_B T} \tag{S4}$$

where V_p is the volume of one particle. By assuming MNPs concentration of 100 mg/L (highest MNPs concentration employed in this experiment), aggregation parameter is calculation as follows:

$$\Gamma = \frac{(4\pi \times 10^{-7})(1.474 \times 10^{5})^{2}(1.414 \times 10^{-23})}{12(1.381 \times 10^{-23})(300)}$$

$$= 7.765$$

$$\phi_{o} = \frac{c}{\rho_{p}}$$

$$= \frac{0.1}{3455.43}$$

$$= 2.894 \times 10^{-5}$$

$$N^{*} = \sqrt{(2.894 \times 10^{-5})e^{7.765 - 1}} = 0.158 < 1$$

Since N* is less than unity even in the most concentrate MNPs solution used in the experiment, MNPs aggregation and MNPs/MNPs interaction is insignificant in the current study. All the calculation performed so far, involved both magnetic Bjerrum length and aggregation parameter further demonstrates the non-interactive nature of the MNPs system used in this investigation. Therefore, MNPs/MNPs interaction can be safely ignored in the analysis of the experimental result in current manuscript.

S4. JUSTIFICATION FOR ONE DIMENSIONAL MAGNETIC FLUX DENSITY APPROXIMATION

In reality, magnetic field strength generated by a cylindrical magnet is a complicated function of x, y and z coordinates in three-dimensional space. However, in order to simplify the model and reduce the computational effort, approximation was made such that magnetic flux density is only the function of axial distance (Equation (7)) in the current work. The three dimensional profile of magnetic flux density is calculated by using COMSOL Multiphysics to justify the suitability of this approximation. As illustrated in Figure S2, the magnetic flux density along the axis of the cylindrical magnet is matching nicely with Equation (7). The magnetic flux density decays rapidly as one moves away from the magnet pole along the axis.



Figure S2. Magnetic flux density profile along the axis of the cylindrical magnet. The solid line represents magnetic flux density calculated by COMSOL Multiphysics in three dimensional space. In contrast, the circular dots depict the magnetic flux density calculated by Equation (7).

On the contrary, according to Figure S3, magnetic flux density does not varies significantly along the radial direction (x and z) of the magnet on a fixed elevation (constant y). Therefore, magnetic flux density is strongly dependent on y but only weakly dependent on x and z. Here, the following approximations were made to simplify the calculation:

$$B(x,y,z) \approx B(0,y,0) \tag{S5}$$

$$\frac{\partial B}{\partial x} \approx 0 \qquad \frac{\partial B}{\partial z} \approx 0$$
 (S6)

$$\nabla B = \frac{\partial B}{\partial x}\vec{e_x} + \frac{\partial B}{\partial y}\vec{e_y} + \frac{\partial B}{\partial z}\vec{e_z} \approx \frac{\partial B}{\partial y}\vec{e_y}$$
(S7)



Figure S3. Magnetic flux density profile along straight lines parallel to the radial direction of the magnet on fixed elevation (constant y). Here, the straight lines span from x = -0.5 cm to x = 0.5 cm and have length of 1 cm (the width of the cuvette where MNPs solution was filled). Due to geometry symmetry, the magnetic flux density profile along the z axis is identical to this image figure.

S5. NON MNPs/FLUID INTERACTING MAGNETOPHORESIS MODEL

Initial and boundary condition. It is assumed that there is no flow (u = 0) and MNPs concentration is uniform throughout the whole solution in the beginning of magnetophoresis. At the bottom boundary of the MNPs solution (the wall adjacent to the magnet), MNPs outlet flux is set as the convective flux due to magnetophoretic migration (= uc) to represent the capture of MNPs which leads to the withdrawal of MNPs from the solution. The other boundaries are assumed rigid and hence there is no MNPs flowing across these boundaries.

Numerical simulation. COMSOL Multiphysics (Version 4.4) was used to solve Equation (2) by utilizing Chemical Reaction Engineering Module in two dimensional space. Physics involved in solving the model mentioned above is 'Transport of Diluted Species' which describes the diffusion and convection of chemical species (which is MNPs in current case) in the solution. A 1×3 cm rectangle was constructed to represent the MNPs solution filled in the disposable cuvette which is similar to the experimental setup. The rectangular domain was then fulfilled with 2128 triangular meshes and 134 boundary elements (by using extra fine element size under physics-controlled mesh) (Figure S4). The result generated was not much different when the mesh is refined, which indicates that the meshes are sufficient to provide an accurate result in this model simulation. Time dependent solver was used to solve the model to predict the transient behavior of MNPs solution under magnetophoresis process. The time span of the simulation was 0 to 240,000 s (4000 minutes) with step size of 1 s. Absolute tolerance of MNPs concentration was set as 0.001 in this simulation. The simulation was done by Dell Precision T3600 Chassis with Intel(R) Xeon(R) Processor E5-1660 (Six Core). The workstation took about 2 hours to complete the simulation.



Figure S4. Mesh element generated by COMSOL Multiphysics in a domain used to represent MNPs solution filled in the disposable cuvette to solve non MNPs/Fluid interacting magnetophoresis model.

S6. HYDRODYNAMICALLY INTERACTING MAGNETOPHORESIS MODEL

Initial and boundary condition. It is assumed that the fluid is stagnant (u = 0) and MNPs concentration is uniform throughout the whole solution in the beginning of magnetophoresis. No slip boundary condition is applied along the boundaries of MNPs solution (cuvette wall) where the fluid velocity is always zero (u = 0). Besides, the pressure at the MNPs solution surface (y = 3 cm) is set equal to atmospheric pressure. All boundaries (except bottom boundary) are assumed rigid and hence there is no MNPs flow across these boundaries. Last but not least, it is necessary to define MNPs outlet flux (which is MNPs capture or separation rate divided by the cross sectional area) at the bottom boundary. In conjunction with that, we

found that the concentration of the MNPs solution decays exponentially with time under an external magnetic field and obeys the following equation up to great precision:

$$c = c_0 e^{-kt}$$
(S8)

$$\ln c = -kt + \ln c_0 \tag{S9}$$

This can be proved by plotting a graph of $\ln c$ versus t, as illustrated in Figure S5. The result demonstrates a good fit of the data into a linear equation (Equation (S9)) which passes through the origin with the coefficient of regression R² is greater than 0.99 and k = 0.001520 min⁻¹ (When normalized concentration is used, $c_0 = 1$ and $\ln c_0 = 0$ which renders the straight line passes through the origin). This has clearly illustrated that the concentration profile is obeying the first order kinetic (Equation (S8)). However, $\ln c$ starts to deviate from the linear line and scatter around after 3000 minutes. This phenomena is due to the limitation of the UV-vis spectrophotometer in detecting the low concentration of MNPs solution in this period of experiment.



Figure S5. The plot of ln ^c against ^t. A straight line passing through the origin can be matched into the plot with coefficient of regression $R^2 = 0.9926$. The gradient of the straight line is given by k = 0.001520 min⁻¹.

Since the MNPs solution is uniformly distributed and amount of MNPs in the solution N is related to MNPs concentration by the equation $N = cV_s$ (where V_s is the volume of MNPs solution), N is also following the first order kinetic with exactly the same rate constant k as that of c:

$$N = c_0 V_s e^{-kt}$$
(S10)

On the other hand, the MNPs outlet flux J at the bottom boundary is given by the derivative of N with respect to time and then divided by the cross sectional area A_b :

$$J = \frac{1 dN}{A_b dt} = \frac{c_0 V_s k}{A_b} e^{-kt}$$
(S11)

Since all parameters $({}^{c_0, V_s, A_b, k})$ in the last equation are known, the MNPs outlet flux at the bottom boundary is now defined. Similar to the concentration of MNPs solution, MNPs outlet flux (and hence MNPs separation rate) also decays exponentially with time.

Numerical simulation. Computational Fluid Dynamics (CFD) and Chemical Reaction Engineering Modules in COMSOL Multiphysics (version 4.4) were employed in the computational work to solve this model in two dimensional space. Continuity and Navier-Stokes equations (Equations (17) and (18)) are found in 'Laminar Flow' physics while driftdiffusion equation (Equation (2)) is contained in 'Transport of Diluted Species' physics. Similar to the previous model, a 1×3 cm rectangle was constructed to represent the MNPs solution filled in the disposable cuvette. The rectangular domain was then filled with 3874 quadrilateral meshes and 230 boundary elements (Figure S6). Time-dependent solver was employed to solve the model in the time span of 0 to 240,000 second (4000 minutes) and time step as long as 10 second was adopted. Due to the highly non-linearity contributed by the magnetophoresis induced convection, the model solution was not able to converge under default solver setting. This problem was solved by manually assigned the scaling factors to some critical dependent variables in this model (MNPs concentration, $c = 3.4 \times 10^{-7}$ mol/m³; pressure, $p = 1 \times 10^5$ Pa and components of velocity field, $u_x \& u_y = 1 \times 10^{-5}$ m/s). The absolute tolerance for MNPs concentration to stop the iteration was set as 0.0005. The simulation was done by Dell Precision T3600 Chassis with Intel(R) Xeon(R) Processor E5-1660 (Six Core). The workstation took about 13 hours to complete the simulation.



Figure S6. Mesh element generated by COMSOL Multiphysics in a domain used to represent MNPs solution filled in the disposable cuvette to solve hydrodynamically interacting magnetophoresis model.

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Symbols	Description	Unit
A	Light absorbance of MNPs solution	-
A_b	Cross sectional area of the cuvette	m ²
В	Magnetic flux density	Т
B _r	Remanent magnetic flux density	Т
β	Volume expansion coefficient	K-1
С	MNPs concentration	kg/m ³
D	Diffusivity of MNPs	m ² /s
d_p	Interparticle distance	m
З	Absorptivity of MNPs	m²/kg
F_d	Viscous drag force	Ν
F_{g}	Gravitational force	Ν
F _{mag}	Magnetic force	Ν
f_m	Volumetric magnetic force on MNPs solution	N/m ³
g	Acceleration due to gravity	m/s^2
Н	Magnetic field strength	A/m
h	Height of cylindrical magnet	m
η	Dynamic viscosity of fluid	N s/m ²
J	MNPs outlet flux	m ⁻² s ⁻¹

k	Rate constant of separation kinetic profile	min ⁻¹
k_B	Boltzmann's constant	J/K
L_c	Characteristic length	m
l	Optical path length	m
λ_B	Magnetic Bjerrum length	m
М	Volumetric magnetization of MNPs solution	A/m
$M_{p,m}$	MNPs mass magnetization	A m²/kg
$M_{p,v}$	MNPs volumetric magnetization	A/m
M _s	Saturation magnetization of MNPs (per unit mass)	A m²/kg
m	Magnetic moment of one magnetic dipole	J m A ⁻¹
m_p	Mass of MNP	kg
Ν	Number of MNPs	-
N^{*}	Aggregation parameter	-
Р	Pressure	N/m ²
ρ	Density of MNPs solution	kg/m ³
$ ho_p$	MNPs density	kg/m ³
R	MNPs core radius	m
R _h	MNPs hydrodynamic radius	m
r	Radius of cylindrical magnet	m
Т	Temperature	Κ
T _s	Heating plate surface temperature	K
T_{∞}	Bulk fluid temperature	K
Г	Magnetic coupling parameter	-
u	Magnetophoretic velocity of MNPs /Velocity field of MNPs	m/s

solution

u_x	<i>x</i> -component of <i>u</i>	m/s
u_y	y-component of u	m/s
u _z	<i>z</i> -component of <i>u</i>	m/s
μ	Magnetic dipole moment	A m ²
μ_0	Permeability of free space	H/m
V	Volume per unit mass	m ³ /kg
V _p	Volume of MNP	m ³
V _s	Volume of MNPs solution	m ³
v	Kinematic viscosity of fluid	m²/s
у	Vertical distance measured from magnet pole	m
ϕ_o	Volume fraction of MNPs in MNPs solution	-

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