Electronic Supplementary Material (ESI) for Lab on a Chip.
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Supporting Information for
A continuum model for magnetic particle flows in microfluidics applicable from dilute to packed suspensions

Supporting notes

Note S1: Drag force in the dilute regime:

The Wen and Yu model (1) is implemented by default in COMSOL for dilute regimes. This formulation is derived from Stokes drag law.

$$F_{d\to c} = -F_{c\to d} = \beta(u_d - u_c)$$

The drag force coefficient writes as

$$\beta = \frac{3\varepsilon_c\varepsilon_d\rho_c C_D}{4d_p}|u_d - u_c|\varepsilon_c^{-2.65}$$

The particle drag coefficient C_D is calculated as follows, using the particle Reynolds number Re_p.

$$Re_p = \frac{2r_p\rho_c|u_d - u_c|}{\mu_c}$$

$$C_D = \begin{cases} \frac{24}{Re_p}\left(1 + 0.15Re_p^{0.687}\right) & Re_p < 1000 \\ 0.44 & Re_p > 1000 \end{cases}$$

Note S2: Drag force in the column regime:

The model introduced here uses a tensor form of the permeability. The interphase drag force is expressed as

$$F_{d\to c} = -F_{c\to d} = \frac{\mu_c\varepsilon_c^2}{K}(u_d - u_c)$$

Which develops as

$$F_{d\to c} = -F_{c\to d} = \mu\varepsilon_c^2\begin{bmatrix} K_{xx}^{-1}(u_{d,x} - u_{c,x}) + K_{xy}^{-1}(u_{d,y} - u_{c,y}) + K_{xz}^{-1}(u_{d,z} - u_{c,z}) \\ K_{yx}^{-1}(u_{d,x} - u_{c,x}) + K_{yy}^{-1}(u_{d,y} - u_{c,y}) + K_{yz}^{-1}(u_{d,z} - u_{c,z}) \\ K_{zx}^{-1}(u_{d,x} - u_{c,x}) + K_{zy}^{-1}(u_{d,y} - u_{c,y}) + K_{zz}^{-1}(u_{d,z} - u_{c,z}) \end{bmatrix}$$

The tensor K is expressed as

$$K = RK_{ref}R^T \qquad with \qquad K_{ref} = \begin{bmatrix} K_{\parallel} & 0 & 0 \\ 0 & K_{\perp} & 0 \\ 0 & 0 & K_{\perp} \end{bmatrix}$$

The components $K_{\parallel}$ and $K_{\perp}$ are given by the model of Whesthuizen and Du Plessis (Eq. 8).

Note S3: Rotation matrix components in 2D

R is the rotation matrix to turn K_{ref} in the direction of the magnetic field H. It is expressed as a function of the angle θ between the magnetic field and the x-axis.

$$R = \begin{bmatrix} \cos\theta & -\sin\theta \\ \sin\theta & \cos\theta \end{bmatrix}$$

With $\cos\theta = H_x/H$ and $\sin\theta = H_y/H$ where $H = \sqrt{H_x^2 + H_y^2 + H_z^2}$ is the magnetic field norm.

The elements of the matrix are

- $R_{11} = H_x/H$
- $R_{22} = H_x/H$
- $R_{12} = H_y/H$
- $R_{21} = -H_y/H$

Note S4: Rotation matrix components in 3D

The rotation matrix in obtained by rotating the reference axis system around an axis **n** with an angle θ. The axis and angle are expressed as functions of the magnetic field components.

The axis of rotation is expressed as

$$\mathbf{n} = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \times \begin{pmatrix} H_x/H \\ H_y/H \\ H_z/H \end{pmatrix} = \begin{pmatrix} 0 \\ -H_z/H \\ H_y/H \end{pmatrix}$$

Angle of rotation (angle between x and H)

$$\cos\theta = \frac{x \cdot H}{|x||H|} = \frac{H_x}{H}$$

Matrix of rotation

$$R = \begin{bmatrix} n_x^2(1-c)+c & n_x n_y(1-c) - n_z s & n_x n_z(1-c) + n_y s \\ n_x n_y(1-c) + n_z s & n_y^2(1-c)+c & n_y n_z(1-c) - n_x s \\ n_x n_z(1-c) - n_y s & n_y n_z(1-c) + n_x s & n_z^2(1-c)+c \end{bmatrix}$$

where
$c = \cos\theta = H_x/H$
$s = \sin\theta = \sqrt{1 - H_x^2/H^2}$

Finally, the matrix elements are expressed without trigonometric functions as

- $R_{11} = H_x/H$
- $R_{12} = -R_{21} = -\frac{H_y}{H}\sqrt{1 - H_x^2/H^2}$
- $R_{13} = -R_{31} = -\frac{H_z}{H}\sqrt{1 - H_x^2/H^2}$
- $R_{22} = \frac{H_z^2}{H^2}\left(1 - \frac{H_x}{H}\right) + \frac{H_x}{H}$
- $R_{23} = R_{32} = -\frac{H_z H_y}{H^2}\left(1 - \frac{H_x}{H}\right)$

- $R_{33} = \frac{H_y^2}{H^2}\left(1 - \frac{H_x}{H}\right) + \frac{H_x}{H}$

Note S5: Implementation of the model in COMSOL

The CFD and AC/DC modules are required to implement this model, we use version 5.5 of COMSOL Multiphysics, but it may work using other versions. This short tutorial is aimed at advanced COMSOL users, who are already familiar with the following notions:

- Laminar flow simulation (e.g. spf module)
- Magnetic field simulation (mf or mfnc modules)
- Manual mesh creation and mesh refinement
- Stationary and Time dependent studies.
- Results visualization and processing in COMSOL.

1. **Create a new model** (2D or 3D) and draw/import the geometry. Create the mesh, and make sure that it is fine where the particle density is expected to be high.

2. **Define the external magnetic field.** It can be done analytically or by simulation.

    a. To define the magnetic field analytically, create analytic functions under the "Definitions" title. For example, in our magnetophoresis example, we define the following functions (see Figure S2):

Name	Arguments	Unit	Expression
Hx	x,y (m)	A/m	Id*x*y/(pi*sqrt(x^2+y^2)^4)
Hy	x,y (m)	A/m	Id/(2*pi*sqrt(x^2+y^2)^2)-Id*x^2/(pi*sqrt(x^2+y^2)^4)
normH	x,y (m)	A/m^2	Id/(2*pi*(x^2+y^2))
normB	x,y (m)	A/m^2	mu0_const*sqrt(Hx(x,y)^2+Hy(x,y)^2)

    b. The magnetic field can also be computed using the **mfnc** or **mf** module. For example, in our magnetic tweezers example, we use the mfnc module. The external permanent magnet is defined in the material section by using 'N50 (Sintered NdFeB)' and in the mfnc section with 'Magnetic Flux Conservation', using 'Remanent flux density' as the Magnetization model. The properties are then taken from material. The magnetic tweezers are defined in a similar way using 'Soft Iron (witout losses)' as a material, which was modified with a custom BH curve (Figure S6). The field is then computed in a separate stationary study.

3. **Define magnetic particle properties** by using custom parameters. For example, with MyOne beads:

Name	Value
d_beads	1[um]
rho_beads	1700[kg/m^3]
Ms	21.86[A*m^2/kg]
gamma	6.28e-5[m/A]

And define an analytical function for the magnetization. (Figure S1)

Name	Arguments	Unit	Expression
massMag	H (A/m)	A*m^2/kg	Ms*(coth(H*gamma)-1/(H*gamma))

4. **Define physics** by adding an Euler-Euler (**ee** module) physics. This physics should be applied only to the fluidic geometry, and not the surrounding volume which may have been necessary for the previous computation of the magnetic field. In the 'Phase Properties' menu, set the required constants for continuous phase density and viscosity (1000[kg/m^3] and 1e-3[Pa*s] for water). For the dispersed phase properties, enter rho_beads and d_beads. For the viscosity model, enter a user defined viscosity with the Einstein formula: ee.muc*(1+2.5*ee.phidReg). Use Pure phase value for the continuous phase and mixture viscosity for the dispersed phase. For Drag and Solid Pressure models, use Gidaspow.

5. **Define boundary and initial conditions** as you need. Particles can be introduced either by defining a volume fraction phid > 0 as initial values in the desired area, or using a volume fraction at the inlet.

6. **Add shallow channel approximation** for 2D Hele-Shaw models. Add a custom volume force as follows (example for 50 μm chamber thickness):

Fc_x	-12*ee.mucm*ucx/(50[μm])^2
Fc_y	-12*ee.mucm*ucy/(50[μm])^2
Fd_x	-12*ee.mucm*udx/(50[μm])^2
Fd_y	-12*ee.mucm*udy/(50[μm])^2

7. **Add magnetic force** by adding a custom volume force defined as follows (Note S4):

Fc_x,y,z	0
Fd_x	if(mfnc.normH>0,mu0_const*rho_beads*massMag(mfnc.normH)*d(mfnc.normH,x),0)
Fd_y	if(mfnc.normH>0,mu0_const*rho_beads*massMag(mfnc.normH)*d(mfnc.normH,y),0)
Fd_z	if(mfnc.normH>0,mu0_const*rho_beads*massMag(mfnc.normH)*d(mfnc.normH,z),0)

Depending on how the magnetic field was defined, the 'mfnc' can be replaced by a custom variable (e.g. mfnc.Bx becomes mu0_const*Hx)

8. **Add the modified drag model**. You will need to activate the 'Equation View' mode (click on the eye icon to see the options).

    a. Define custom parameters for permeability as follows:

Kpara	(pi+2.157*ee.phidReg)*(1-ee.phidReg)^2/(48*ee.phidPos^2)*rp^2
Kperp	pi*(1-ee.phidReg)*(1-sqrt(ee.phidReg))^2/(24*ee.phidPos^(3/2))*rp^2

    b. Define the reference matrix named 'Kref' as:

Kpara	0	0
0	Kperp	0
0	0	Kperp

    c. Define the rotation matrix named 'R' as (Note S4):

mfnc.Bx/mfnc.normB	-mfnc.By/mfnc.normB *sqrt(1-mfnc.Bx^2/mfnc.normB^2)	mfnc.Bz/mfnc.normB*sqrt(1-mfnc.Bx^2/mfnc.normB^2)
mfnc.By/mfnc.normB*sqrt(1-mfnc.Bx^2/mfnc.normB^2)	mfnc.Bz^2/mfnc.normB^2*(1-mfnc.Bx/mfnc.normB)+mfnc.Bx/mfnc.normB	mfnc.Bz*mfnc.By/mfnc.normB^2*(1-mfnc.Bx/mfnc.normB)
-mfnc.Bz/mfnc.normB*sqrt(1-mfnc.Bx^2/mfnc.normB^2)	mfnc.Bz*mfnc.By/mfnc.normB^2*(1-mfnc.Bx/mfnc.normB)	mfnc.By^2/mfnc.normB^2*(1-mfnc.Bx/mfnc.normB)+mfnc.Bx/mfnc.normB

    As before, depending on how the magnetic field was defined, the 'mfnc' can be replaced by a custom variable (e.g. mfnc.Bx becomes mu0_const*Hx)

    d. Define the transposed rotation matrix named 'Rt' as:

R11	R21	R31
R12	R22	R32
R13	R23	R33

e. Perform a first matrix product R*Kref by defining a matrix named 'RKref' as follows:

Kref11*R11+Kref21*R12+Kref31*R13	Kref12*R11+Kref22*R12+Kref32*R13	Kref13*R11+Kref23*R12+Kref33*R13
Kref11*R21+Kref21*R22+Kref31*R23	Kref12*R21+Kref22*R22+Kref32*R23	Kref13*R21+Kref23*R22+Kref33*R23
Kref11*R31+Kref21*R32+Kref31*R33	Kref12*R31+Kref22*R32+Kref32*R33	Kref13*R31+Kref23*R32+Kref33*R33

f. Perform a second matrix product to obtain the final permeability matrix K = R*Kref*Rt. Define a matrix named 'RKrefRt' as follows:

Rt11*RKref11+Rt21*RKref12+Rt31*RKref13	Rt12*RKref11+Rt22*RKref12+Rt32*RKref13	Rt13*RKref11+Rt23*RKref12+Rt33*RKref13
Rt11*RKref21+Rt21*RKref22+Rt31*RKref23	Rt12*RKref21+Rt22*RKref22+Rt32*RKref23	Rt13*RKref21+Rt23*RKref22+Rt33*RKref23
Rt11*RKref31+Rt21*RKref32+Rt31*RKref33	Rt12*RKref31+Rt22*RKref32+Rt32*RKref33	Rt13*RKref31+Rt23*RKref32+Rt33*RKref33

g. Inverse the permeability matrix by using the inversion tool of COMSOL. Define an inverse matrix named 'invK' with the following components:

RKrefRt11	RKrefRt12	RKrefRt13
RKrefRt21	RKrefRt22	RKrefRt23
RKrefRt31	RKrefRt32	RKrefRt33

h. Incorporate the permeability matrix into drag model (Equations 4-8). Under 'Phase Properties', go in 'Equation View'. Six components have to be replaced as follows (see Note S2):

ee.Inter_force_dx	nojac(gpeval(5,ee.muc*ee.phicReg/ee.phidPos,0))*(nojac(gpeval(5,invK.invT11,0))*ee.uslipx+nojac(gpeval(5,invK.invT12,0))*ee.uslipy+nojac(gpeval(5,invK.invT13,0))*ee.uslipz)
ee.Inter_force_dy	nojac(gpeval(5,ee.muc*ee.phicReg/ee.phidPos,0))*(nojac(gpeval(5,invK.invT21,0))*ee.uslipx+nojac(gpeval(5,invK.invT22,0))*ee.uslipy+nojac(gpeval(5,invK.invT23,0))*ee.uslipz)
ee.Inter_force_dz	nojac(gpeval(5,ee.muc*ee.phicReg/ee.phidPos,0))*(nojac(gpeval(5,invK.invT31,0))*ee.uslipx+nojac(gpeval(5,invK.invT32,0))*ee.uslipy+nojac(gpeval(5,invK.invT33,0))*ee.uslipz)
ee.Inter_force_cx	nojac(gpeval(5,ee.muc,0))*(nojac(gpeval(5,invK.invT11,0))*ee.uslipx+nojac(gpeval(5,invK.invT12,0))*ee.uslipy+nojac(gpeval(5,invK.invT13,0))*ee.uslipz)
ee.Inter_force_cy	nojac(gpeval(5,ee.muc,0))*(nojac(gpeval(5,invK.invT21,0))*ee.uslipx+nojac(gpeval(5,invK.invT22,0))*ee.uslipy+nojac(gpeval(5,invK.invT23,0))*ee.uslipz)
ee.Inter_force_cz	nojac(gpeval(5,ee.muc,0))*(nojac(gpeval(5,invK.invT31,0))*ee.uslipx+nojac(gpeval(5,invK.invT32,0))*ee.uslipy+nojac(gpeval(5,invK.invT33,0))*ee.uslipz)

9. **Run a time dependent study** for the Euler-Euler physics. If the magnetic field was computed in a previous study, you have to specify it in 'Values of variables not solved for' in the time dependent study parameters. For more stability at the first steps of computation, it can be worth the pre-calculate an initial flow field using a separate laminar flow physics (spf module), and specify its results in 'Initial values of variable solved for'.

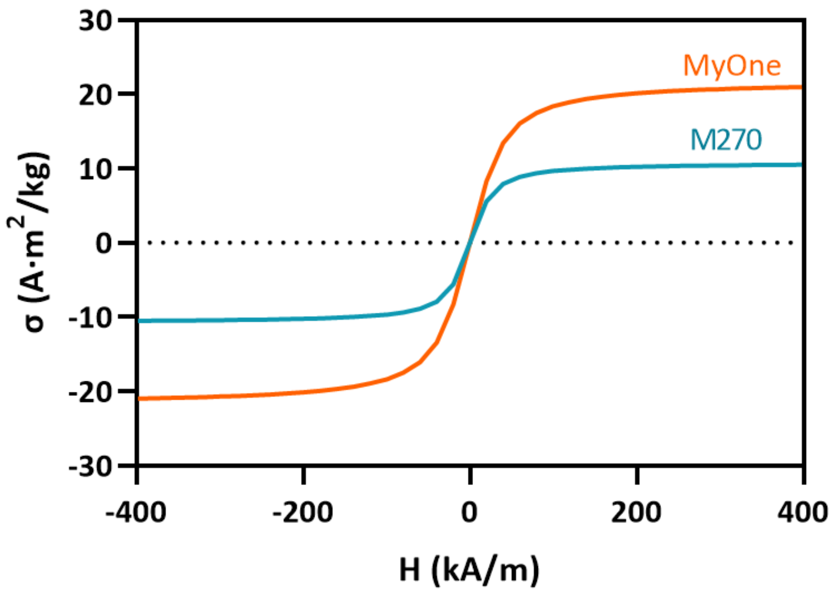

Figure S1: Magnetization curves of M270 and MyOne magnetic particles. The magnetization per unit mass σ (A · m^2/kg) as a function of the external magnetic field H (A/m) is obtained by fitting the magnetization curve data provided by the supplier with a Langevin function: $\sigma(H) = M_s\left(\coth(\gamma H) - \frac{1}{\gamma H}\right)$. The fittings give: $M_{s,M270} = 10.8 emu/g,\ \gamma_{M270} = 9.47 \cdot 10^{-5}\, m/A$ and $M_{s,MyOne} = 21.86 emu/g,\ \gamma_{MyOne} = 6.28 \cdot 10^{-5}\, m/A$.

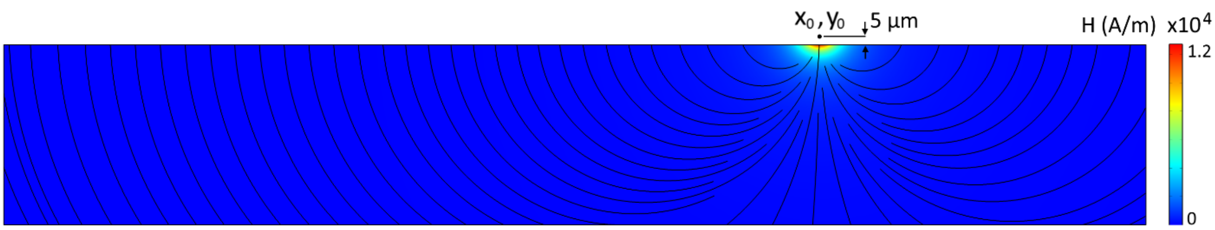

Figure S2: Magnetic field with $id = 12$ µAm. The magnetic field components around the wire write as: $H_x = \frac{idxy}{\pi(x^2 + y^2)}$ and $H_y = -\frac{id}{2\pi(x^2 + y^2)} - \frac{idx^2}{\pi(x^2 + y^2)^2}$. The wire is placed 5 µm vertically from the top boundary, and 250 µm horizontally from the fluid inlet.

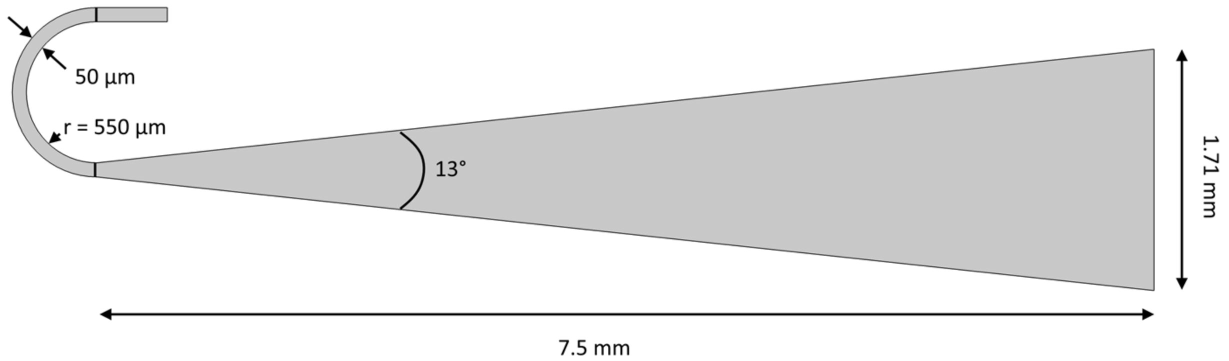

Figure S3: Fluidized bed geometry.

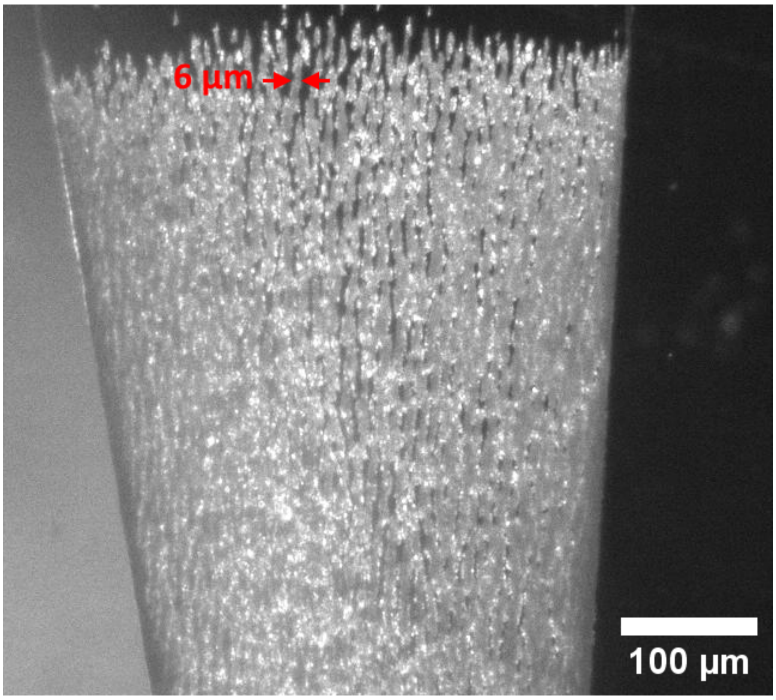

Figure S4: Column-shaped aggregates in the fluidized bed. The column diameter is uneven and usually higher than the particle size (2.8 µm). For the simulation, we took a column diameter of 6 µm, which corresponds to a typical observation.

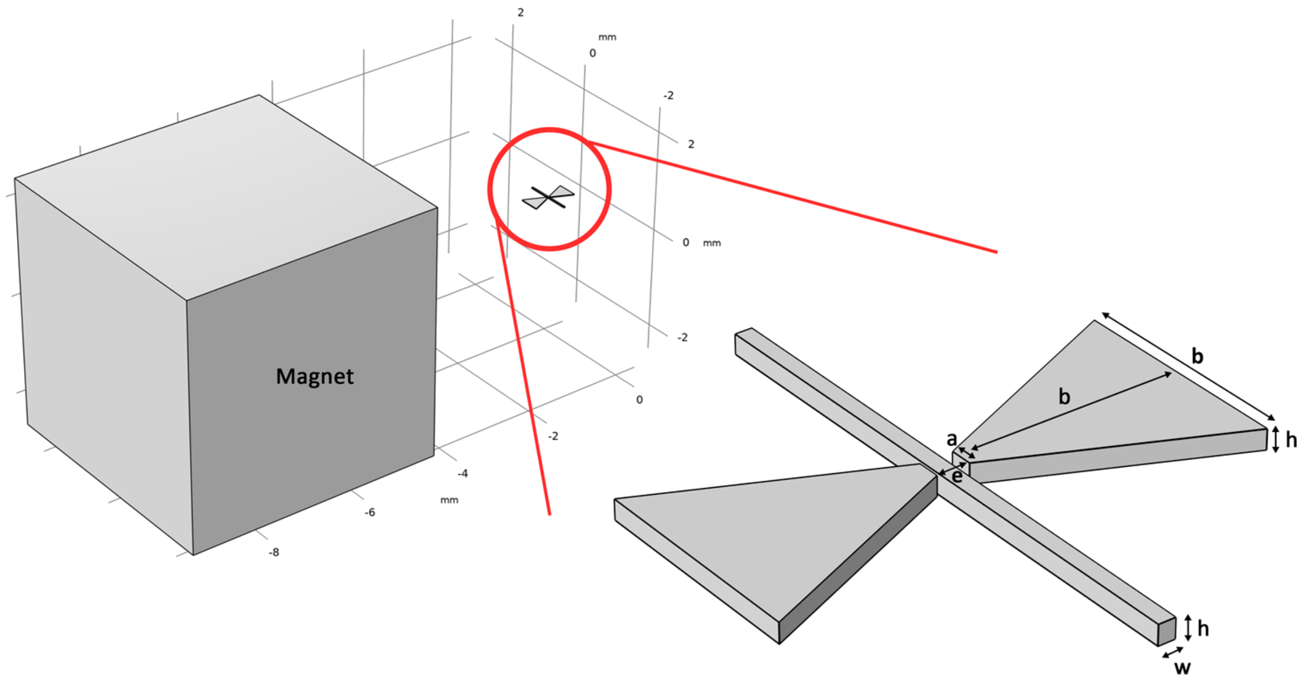

Figure S5: Magnetic microtweezers geometry. Magnet size = 5x5x5 mm, Distance between magnet and tweezers = 4 mm, a (wide) = 40 µm, a (sharp) = 0 µm, b = 400 µm, e = 50 µm, h = 35 µm, w = 30 µm. Channel length = 1 mm.

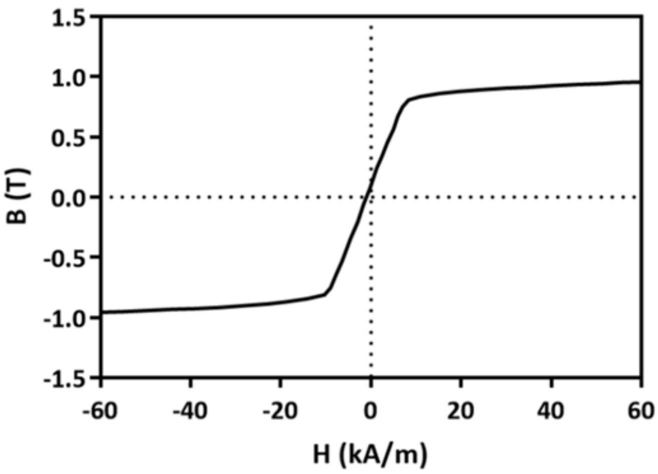

Figure S6: Magnetization curve of the NiFe alloy, from Dumas *et al.* (2) This curve is set as material properties for the tweezers in COMSOL.

Table S1: Bead properties

Used in	Bead reference	Diameter (μm)	Density (kg/m³)	Ms (emu/g = Am²/kg)	$\gamma \times 10^{-5}$ (m/A)
Dilute magnetophoresis and Magnetic microtweezers	Dynabeads MyOne	1	1800	21.86	6.28
Fluidized bed	Dynabeads M270	2.8	1300	10.80	9.47

Table S2: Grid convergence study

a) Magnetophoresis in the dilute regime

	Av. element size in the cluster region (μm)	Computation time (h)	Cluster size (um²)	Variation %
Very coarse	0.8	0.06	6.70	
Coarse	0.6	0.3	8.66	29.3%
Medium	0.4	1.06	10.52	21.4%
Fine	0.3	2.33	11.36	7.9%
Very fine	0.2	5.93	11.69	2.9%

b) Microfluidic magnetic fluidized bed

	Av. element size (μm)	Computation time (h)	Bed area	Variation %
Very coarse	37.71	0.35	3.88	
Coarse	25.14	1.25	3.76	3.19%
Medium coarse	15.08	1.40	3.57	4.94%
Medium	12.57	1.78	3.52	1.42%
Medium fine	10.77	2.4	3.49	0.90%
Fine	9.43	2.83	3.46	0.76%
Finer	8.38	3.58	3.45	0.49%
Very fine	7.54	4.75	3.43	0.45%

c) Magnetic microtweezers

	Av. element size in the cluster region (μm)	Computation time (h)	Cluster size (ng)	Variation %
Very coarse	3.78	0.58	12.23	
Coarse	3.24	0.8	12.55	2.63%
Medium	2.71	1.45	13.85	10.32%
Fine	2.16	2.5	14.68	6.01%
Finer	1.92	4.22	15.04	2.45%
Very fine	1.82	4.77	15.11	0.47%

SI References

1. COMSOL®, CFD Module User's Guide. *COMSOL Multiphysics*, 1–710 (2016).
2. S. Dumas, M. Richerd, M. Serra, S. Descroix, Magnetic Microtweezers: A Tool for High-Throughput Bioseparation in Sub-Nanoliter Droplets. *Adv. Mater. Technol.* **8**, 2200747 (2023).