Supplemental Information: Magnetically Driven Lipid Vesicles for Directed Motion and Light-Triggered Cargo Release

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Simulation Methods

Magnetic field between two magnets. We obtained the magnetic field density between the two magnets placed in oppositely polarized fashion using *Magnetic Fields, No Current* module of COMSOL Multiphysics software. We modeled the experimental setup in 2D axis-symmetric geometry. The magnets were modeled as two rectangular domains each 0.5 inches wide and 1 inch long separated by a distance of 55 mm. A bigger rectangular domain 5 inches in width and 9.6 inches in length was considered, including both magnets. We defined the remanent flux density of 13 200 G for each magnets in opposite directions. We used the free triangular mesh to discretize all of the domains and solve for the magnetic field using MUMPS solver.

Lattice Boltzmann method. We utilized the lattice Boltzmann method (LBM), a mesoscopic numerical technique, to resolve the hydrodynamics. The fundamental element of the LBM is the discrete-velocity distribution function $n_i(\boldsymbol{r},t)$, which shows the density of fictitious fluid particles moving with a discrete velocity $\boldsymbol{c}_i$ at position $\boldsymbol{r}$ and time t. The fictitious fluid particles only stream with a set of q velocities $\{\boldsymbol{c}_i\}$, where $i=0,...,q-1$, on a regular lattice with sufficient symmetry such that the conservation laws are fulfilled and the macroscopic hydrodynamic fields are recovered. We applied the D3Q19 model, which indicates the fluid particles stream along 19 directions in a 3-D space. The evolution of the distribution function is governed by the lattice Boltzmann equation

$$n_i(\boldsymbol{r}+\boldsymbol{c}_i\Delta t, t+\Delta t) = n_i(\boldsymbol{r},t) + \sum_{i=0}^{19} \mathcal{L}_{ij} n_j^{\mathrm{neq}}(\boldsymbol{r},t) + n_i'(\boldsymbol{r},t), \tag{S1}$$

where $\boldsymbol{r}$ denotes the position vector of the lattice site, t is the time, Δt is the discrete time step, $n_j^{\mathrm{neq}} \equiv n_j - n_j^{\mathrm{eq}}$, and $\mathcal{L}_{ij}$ represents the matrix element of the collision operator. Here, a multi-relaxation time scheme was employed for the collision operator [1]. The disturbance to n_i from the external body force is incorporated by $n_i'(\boldsymbol{r},t)$. The equilibrium distribution function n_i^{eq} for the D3Q19 model is often chosen as

$$n_i^{\mathrm{eq}} = w_i \left(\rho + \frac{\boldsymbol{j}\cdot\boldsymbol{c}_i}{c_s^2} + \frac{\rho \boldsymbol{u}\boldsymbol{u} : (\boldsymbol{c}_i\boldsymbol{c}_i - c_s^2\mathbf{1})}{2c_s^4} \right), \tag{S2}$$

where w_i, is the weight factor for the D3Q19 model, ρ is the fluid density, $\boldsymbol{u}$ is the fluid velocity, the momentum flux is written as $\boldsymbol{j}=\rho\boldsymbol{u}$, and $c_s = c/\sqrt{3}$ with $c = \Delta x/\Delta t$. The lattice spacing Δx and the time step Δt are often set to unity. Two eigenvalues of the collision operator λ and λ_b control the shear and bulk viscosities

$$\mu = -\rho c_s^2 \Delta t \left(\frac{1}{\lambda_s} + \frac{1}{2} \right), \quad \mu_b = -\rho c_s^2 \Delta t \left(\frac{2}{3\lambda_b} + \frac{1}{3} \right), \tag{S3}$$

where μ and μ_b denote the shear and bulk viscosities, respectively [1]. Other macroscopic quantities, such as ρ and $\boldsymbol{j}$, can be derived from the weighted sum of the density distribution function as follows:

$$\rho(\boldsymbol{r},t) = \sum_{i=1}^{19} n_i(\boldsymbol{r},t), \tag{S4}$$

$$\boldsymbol{j}(\boldsymbol{r},t) = \sum_{i=1}^{19} n_i \boldsymbol{c}_i + \frac{1}{2}\boldsymbol{f}(\boldsymbol{r},t)\Delta t, \tag{S5}$$

where $\boldsymbol{f}(\boldsymbol{r},t)$ is the external body force acting on the fluids. In this study, it is the restoring force of the membrane as detailed below.

Solid particle model. We employed the simple bounce-back scheme [1] to enforce the no-slip condition on the translating sphere and the solid walls that enclose the simulation domain. The momentum exchange between fluids and the solid sphere can be computed via the simple bounce-back scheme. The sphere positions were updated by integrating its equations of motion using an implicit scheme [2].

Membrane model. The membrane surface was discretized by a network composed of N_v vertexes, N_b bonds, and N_f triangular faces, where $N_v = 2562$, $N_b = 7680$, and $N_f = 5120$. The bending resistance was modeled by the bending energy

$$E_b = \sum_{i=1}^{N_b} k_b \left[1 - \cos(\theta_i - \theta_0)\right], \tag{S6}$$

where k_b is the model bending constant, θ_i and θ_0 are the dynamic and rest dihedral angles between two triangular patches on the discretized membrane surface, respectively. The relation between k_b and the bending rigidity k_c of the Helfrich model is given by $k_b = 2k_c/\sqrt{3}$ [3]. To model the area incompressibility of the membrane, we imposed the area conservation constraint as follows:

$$E_a = \frac{k_a(A - A_0)^2}{2A_0} + \sum_{i=1}^{N_f} \frac{k_l(a_i - a_{0,i})^2}{2a_0}, \tag{S7}$$

where a_i and $a_{0,i}$ are the dynamic and rest areas of each triangular face, respectively. Similarly, A and A_0 are the dynamic and rest areas of the membrane, respectively. The constants k_a and k_l control the strength of these area constraint forces. To prevent the enclosed volume drifts from its initial value V_0, we also applied a volume conservation constrain

$$E_v = \frac{k_v(V - V_0)^2}{2V_0}, \tag{S8}$$

where k_v is the constraint strength. V denotes the enclosed volume. In addition to the aforementioned potentials, we also applied a tethering potential E_s on each bond of the discretization network. Therefore, our model resembles an enclosed polymerized membrane. E_s comprises an attraction (E_{att}) and a repulsive (E_{rep}) interactions: $E_s = E_{\text{att}} + E_{\text{rep}}$, with

$$E_{\text{att}} = \sum_{i=1}^{N_b} \frac{k_{\text{B}}Tl_m(3\xi_i^2 - 2\xi_i^3)}{4p(1 - \xi_i)}, \tag{S9}$$

where $\xi_i = l_i/l_m$ with the bond length l and the maximum bond length l_m, $0 < \xi_i < 1$, p is the persistence length, and $k_{\text{B}}T$ is the energy unit, and

$$E_{\text{rep}} = \sum_{j=1}^{N_b} \frac{k_p}{l_j}, \tag{S10}$$

where k_p is a force constant. The relation between these discrete model parameters and the macroscopic properties of this polymerized membrane model can be found in [4]. The total force $\boldsymbol{f}_j$ acting on each membrane vertex is obtained from $\boldsymbol{f}_j = -\frac{\partial E}{\partial \boldsymbol{x}_j}$, where $E = E_b + E_a + E_v + E_s$, and from $\boldsymbol{f}_j$, the external body force $\boldsymbol{f}(\boldsymbol{r}, t)$ can be calculated. The position of the membrane vertex was then updated by the Adams–Bashforth method. The coupling between the membrane and the fluids was realized by the immersed boundary method with a two-point stencil [5].

In the simulation, a constant force of 4.85 pN was applied to the solid sphere based on the predicted magnetic field density. High bending and shear moduli were employed in the simulations to replicate the largely undeformed membrane shapes observed in experiments. The parameters of the membrane model used in the simulations are listed in Table S1.

Symbol	Parameter	Value
k_c	bending constant	1.47×10^{-16} N $\cdot$ m
k_a	global area constraint	4.72×10^{-4} N/m
k_l	local area constraint	4.72×10^{-4} N/m
k_v	volume constraint	9.50×10^{2} N/m^2
G	shear modulus	10^{-4} N/m

TABLE S1. Membrane model parameters. The connection between the macroscopic characteristics and the model parameters can be found in [4].

Supporting Movies

Movie S1. The movie (played at 4X speed) shows the motion of a magGUV containing 1000 mM sucrose and 20 mM of a photosensitizer HPTS in an external isomolar 1020 mM sucrose solution. Following light-triggered asymmetric oxidation, magGUV explodes, releasing the inner contents as well as the encapsulated magnetic microparticle. The scale bar represents 20 μm.

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