SUPPORTING INFORMATION

Metachronal $\mu$-Cilia for On-Chip Integrated Pumps and Climbing Robots

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Other supplementary material for this manuscript includes the following:

Movies S1 to S12
S1. Paramagnetic Particle Distribution Resulting from Different Arrangements of Magnets

Based on the mechanism that paramagnetic particles form structures that tend to align with the applied magnetic field during the sample curing step in the µMAC fabrication process, we simulated three different configurations of the magnets, namely a single magnet with its North Pole facing the mold (Figure S1 A1 to A3), a magnet-array arranged naturally, i.e. with the opposing magnetic poles connected to each other, hence termed “natural configuration” (Figure S1 B1 to B3), and a magnet-array that is arranged with alternating dipole orientation between adjacent magnets, hence termed “staggered configuration” (Figure S1 C1 to C3). The staggered configuration gives the best result in terms of both the generated magnetic field and the resulting magnetic particle alignment. Therefore, we used the staggered configuration to fabricate the metachronal µMAC in the main text.
**Figure S1.** Paramagnetic particle distribution resulting from different arrangements of magnets. (A1 to C1) Snapshot of a COMSOL simulation of the magnetic flux density $B$ induced by a single rod-shaped magnet with its North Pole facing the mold (A1), a
magnet-array arranged with the natural configuration (B1), and a magnet-array arranged with the staggered configuration (C1). The horizontal white lines indicate the central position of the µMAC array, i.e. 0.7 mm above the top surface of the magnet or the magnet-array. The red arrows indicate both the direction and the magnitude of $B$ at the locations of each cilium with a pitch of 350 µm. (A2 to C2) Magnetic field generated by the single rod-shaped magnet (A2), the magnet-array with the natural configuration (B2), and the magnet-array with the staggered configuration (C2) from a COMSOL simulation, along the horizontal µMAC level in panel A1 to C1, respectively. (A3 to C3) Schematic drawing of one row of µMAC (pitch = 350 µm) indicating the arrangement of the paramagnetic particles (black line segments) according to the COMSOL simulation in panel A1 to C1, respectively, by assuming that the particle chain direction is in the same the direction as the magnetic field. The middle of the µMAC array is assumed to be placed at $x = 0$ in panel A1 to C1, respectively. The theoretically predicted acute angle ($\theta$) between the magnetic particle chain and the vertical direction is shown above the corresponding cilium. Illustration is not to scale.
S2. Magnetic Field Generated by the Rotating Magnets

The magnetic field generated by the two magnets in the actuation setup shown in Figure 3A was measured using a gauss meter (F.W. Bell 811A Digital Gaussmeter connected with a Model HTL 4-0608 probe). The magnetic gradient was calculated based on the measured magnetic field.
**Figure S2.** Magnetic field generated by the two rotating magnets shown in Figure 3A.

(A) Schematic diagram of the magnetic actuation setup for creating a 2D rotating magnetic field, indicating the xyz coordinates. \((x, y, z) = (0, 0, 0)\) represents the spatial center. Illustration is not to scale. (B) The measured magnetic field along the central x axis of the space between the two magnets. (C) Calculated magnetic gradient along the central x axis based on the results in panel B. Each data point was obtained by averaging the results of at least five measurements.
Figure S3. Motion of the whole µMAC array and one control cilium at 1 Hz in the 2D rotating magnetic field in both water and glycerol. The blue dashed line and the red dashed line indicate the tip trajectory during the magnetic stroke and elastic stroke, respectively. The µMAC array has a pitch of 550 µm. Note the difference in the recording speed for the metachronal µMAC and the control µMAC, as well as for in water and in glycerol, respectively (see Experimental Section for details).
S4. Trajectories of Tracer Particles in both Water and Glycerol

| 350 µm pitch | 450 µm pitch | 550 µm pitch | 550 µm pitch control |
|--------------|--------------|--------------|-----------------------|
| (12 x 10 = 120 cilia) | (9 x 10 = 90 cilia) | (8 x 10 = 80 cilia) | (8 x 10 = 80 cilia) |

**Figure S4.** Trajectories of tracer particles in both water and glycerol. The arrows indicate the relative speed and direction of the generated flow at 10 Hz. The top row images are overlays of 11 images recorded during 1 s, and the bottom row images are overlays of 6 images recorded during 50 s, made with ImageJ.
S5. Calculation of the Generated Volumetric Flow Rate and Pressure Drop

When the maximum velocity within the microfluidic channel at actuation frequency $f$ is known, the corresponding volumetric flow rate $Q_f$ and the pressure drop $\Delta P_f$ (which is the pressure difference between the locations right before and after the ciliated area) generated by the µMAC can be calculated as follows:\(^{40}\)

$$\Delta P_f = \frac{\eta \nu_x(0, 0)L\pi^2}{4w^2\sum_{i=1,3,5,...}(-1)^{i-1}2\left[1 - \frac{1}{\cosh\left(\frac{im\pi}{2w}\right)}\right]^3}$$  \hspace{1cm} (S1)

$$Q_f = \frac{\Delta P_f hw^3}{12\eta L} \left[1 - \frac{192w}{\pi^3h} \sum_{i=1,3,5,...} \frac{\tanh\left(\frac{in\pi}{2w}\right)}{i^5}\right]$$  \hspace{1cm} (S2)

where $\eta$ is the dynamic viscosity of the liquid at room temperature ($1$ mPa·s for water and $1.4$ Pa·s for glycerol), $\nu_x(0, 0)$ is the measured flow speed in the geometrical center of the channel at the actuation frequency of $f$, $w$ is the width of the channel, $h$ is the height of the channel, $L$ is the length of the channel. In our study, $w = 6$ mm, $L = (10 + 6) \times 4 - 4 = 60$ mm (4 mm is the length of the ciliated area), $h = 2$ mm. The calculated flow rate and pressure drop are depicted in Figure S5.
Figure S5. Corresponding volumetric flow rate (A) and pressure drop (B). Each data point was obtained by averaging the results of at least ten measurements.
Figure S6. Tip speed and swept area of µMAC motion at 10 Hz in both water (A) and glycerol (B).
S7. Maximum Local Reynolds Number (Re\text{max})

The maximum local Reynolds number (Re\text{max}) is defined by Re\text{max} = \rho \nu \ell / \eta, in which \rho is the density of the fluid at room temperature, \nu is the maximal speed of the cilia tips (see Figure 4A and Figure S6A), \ell is the characteristic length for which we take the cilia length (350 \mu m in our experiments), and \eta is the dynamic viscosity of the fluid at room temperature (1 mPa\cdot s for water and 1.4 Pa\cdot s for glycerol). The calculated Re\text{max} is plotted in Figure 4B in both water and glycerol.
S8. Comparison of Fluid Pumping Capability of our Metachronal µMAC with other Artificial Cilia and Microfluidic Pumps

A comparison between our metachronal µMAC (pitch = 350 µm) and previously published artificial cilia is summarized in Table S1. The normalized flow speed by $v/\mu L$ is depicted in Figure S7, where $v$ is the maximum flow speed, $\mu$ is the beating frequency of the cilia, and $L$ is the length of the cilia. It can be concluded that our µMAC outperform all reported artificial cilia in terms of generated global flow speed, except for the µ-cilia reported in refs. 15,26, and the millimetre scaled pneumatic cilia.23

Our metachronal µMAC (pitch = 350 µm) are capable of generating a water volumetric flow rate $Q_{max}$ of 85 µL min$^{-1}$ in the square channel shown in Figure 3B. Comparing the flow rate to those that have been achieved using electro-hydrodynamic and electroosmotic pumping methods,50 our cilia-based pumping is in the upper range, see Figure S8. However, this comparison is for widely different channel sizes, and to account for this, Laser and Santiago have defined the “self-pumping performance”,
$Q_{max}/S_p$ as a parameter that can be used to characterize and compare micropumping efficiencies; $S_p$ is the total volume of the channel.\textsuperscript{50} Our circular channel has a total volume $S_p$ of 768 mm\textsuperscript{3}, which gives a self-pumping performance of 0.11 min\textsuperscript{-1}. Figure S8 shows that our metachronal µMAC is in the upper range of self-pumping performance compared with the other methods of microfluidic pumping reviewed by Laser and Santiago\textsuperscript{50} while the size is one of the smallest.
Table S1. Comparison between our metachronal µMAC (italic) and previously published artificial cilia in terms of fluid pumping in water unless otherwise specified. In the table, these artificial cilia are arranged according to their length from low to high. The best local flow above the ciliated surface is highlighted in bold font, and the best global flow in the channel is marked red.

| Cilia type                  | Cilia length $L$ [μm] | Maximum flow speed $v$ [μm s$^{-1}$] | Beating frequency $f$ [Hz] | Normalized flow speed $v/fL$ |
|----------------------------|-----------------------|-------------------------------------|---------------------------|-----------------------------|
| Electrostatic cilia$^{24a)}$ | 100                   | 600$^{c)}$                           | 200                       | 0.03                        |
| µMAC$^{15}$                | 200                   | 1350$^{d)}$                          | 62                        | 0.11                        |
| µMAC$^{38}$                | 250                   | 120$^{d)}$                           | 20                        | 0.024                       |
| µMAC$^{22}$                | 300                   | 70$^{d)}$                            | 50                        | 0.0047                      |
| µMAC$^{40}$                | 350                   | 250$^{d)}$                           | 40                        | 0.018                       |
| Metachronal-2D µMAC$^{26}$ | 350                   | 3000$^{d)}$                          | 100                       | 0.086                       |
| Our metachronal-2D µMAC    | 350                   | 200$^{d)}$                           | 10                        | 0.063                       |
| Metachronal-2D MAC$^{28b)}$ | 1000                  | 450$^{c)}$                           | 2.5                       | 0.18                        |
| Metachronal-3D MAC$^{19b)}$ | 4000                  | 83$^{c)}$                            | 0.083                     | 0.25                        |
| Pneumatic metachronal-2D cilia$^{23}$ | 8000 | 19000$^{d)}$ | 30 | 0.079 |

$^a$working fluid: silicone oil; $^b$working fluid: glycerol; $^c$maximum local flow above the cilia tips; $^d$maximum global flow in the channel. Note that the normalized flow speed is calculated regardless the chip geometry and the number of cilia.
Figure S7. Comparison of the pumping capability of our metachronal µMAC with previously reported artificial cilia. The normalized flow speed represents $\frac{v}{fL}$ in Table S1. The reference articles are arranged according to their reported cilia length from low to high with ours placed at the rightmost position.
**Figure S8.** Comparison of the pumping capability of our metachronal µMAC with reported pumping methods. (A) The volumetric flow rate compared with electro-hydrodynamic and electroosmotic pumping. (B) Comparison of the self-pumping frequency and size for a range of micropumps; the size of the data point marker
indicates the associated $\Delta P$. Reproduced with permission with our data inserted.\textsuperscript{50}

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S9. Estimated robot locomotion due to wave-like body deformation

Shinoda et al. present magnetic soft robots shaped as rectangular thin films (without cilia or legs) that exhibit net motion due to the undulating wavy deformation of the soft body.\textsuperscript{30} This robot has a length, width and thickness of 8, 1.5 and 0.15 mm respectively and is actuated by a magnetic field rotating at 2.5 Hz. Due to this, the robot performs a sinusoidal wave motion with an amplitude of 0.15 mm, and this results in a net locomotion speed of about 0.15 mm/s.

Our ciliated metachronal robots exhibit similar wavy deformation, as shown in Fig. 5A and B, as well as in movie S7. Our robots are smaller in size than the films of Shinoda et al. (length, width, and thickness of 4, 3.5. and 0.15 mm, respectively),\textsuperscript{30} and the amplitude of the wave is also smaller, namely 0.1 mm (Fig. 5B). Its locomotion speed is nevertheless substantially larger than that of the robot of Shinoda et al.,\textsuperscript{30} namely around 0.7 mm/s (Fig. 7D). This leads us to conclude that the contribution of the wavy
deformation to the total locomotion is likely minor relative to the effect of the metachronal motion of the cilia, acting as the robot’s legs.
S10. Movie Legends

Unless specified, for all movies, the µMAC are made of a magnetic mixture of 2 PDMS : 1 CIP, and for the experiments of ciliated soft robots the µMAC have a pitch of 350 µm.

**Movie S1.** Paramagnetic particle aligning process during µMAC fabrication. The weight ratio of the magnetic particles is 20%, i.e. 4 PDMS : 1 CIP. The µMAC have a pitch of 450 µm. The magnet-array used here is arranged with alternating dipole orientation between neighboring magnets, i.e. the staggered configuration in Figure 1B and Figure S1C. Initially, the magnet-array is sufficiently far away from the mold to ensure the natural magnetic particle distribution not disturbed. Later on, the magnet-array is slowly approaching the mold until it touches the bottom of the mold (~17 s in the movie when the sample is obviously pushed upward). Then the sample is maintained untouched.

**Movie S2.** Bending of one row of µMAC in a 1D uniform magnetic field varying from 0 to 280 mT. The µMAC have a pitch of 350 µm.
Movie S3. Motion of one row of metachronal μMAC in the 2D rotating magnetic field in both water and glycerol at 1 Hz. The μMAC have a pitch of 550 μm.

Movie S4. Motion of one row of control μMAC in the 2D rotating magnetic field in both water and glycerol at 1 Hz. The μMAC have a pitch of 550 μm.

Movie S5. Motion of one row of metachronal μMAC with a pitch of 350 μm in the 2D rotating magnetic field in water at 1 Hz.

Movie S6. Motion of cilium 4 at 1 Hz in the 2D rotating magnetic field during the elastic stroke in both water and glycerol.

Movie S7. Locomotion of metachronal robot and the free motion of one row of metachronal μMAC in air in the 2D rotating magnetic field. The beating frequency of the μMAC is 1 Hz. The blue curve is the trajectory of the robot central body tracked with
ImageJ. The quantitative results of the displacement of the robot central body are depicted in Figure 5B.

**Movie S8.** Bi-directional locomotion of metachronal robot in the 2D rotating magnetic field. The beating frequency of the µMAC is 1 Hz.

**Movie S9.** Locomotion of the control robot in the 2D rotating magnetic field. The beating frequency of the µMAC is 1 Hz.

**Movie S10.** Metachronal robot climbing across a hill. The PMMA hill has a slope angle of 45º. The beating frequency of the µMAC is 2 Hz.

**Movie S11.** Metachronal robot climbing slopes with an angle ranging from 0º to 180º in air. The beating frequency of the µMAC is 2 Hz.
**Movie S12.** Metachronal robot carrying a cargo in both air and liquid. The liquid is pure ethanol. The beating frequency of the μMAC is 2 Hz.