Quantifying the Ethanol Release from the Hydrogel

We calculated the rate of ethanol release from the gel boat by treating it as a cylinder with one end open to mass transfer and uniform initial ethanol concentration (Scheme S1). When the particle is floated on water, the ethanol concentration in the vicinity of the open end of the cylinder is assumed to be zero, because buoyancy carries the released ethanol to the surface and does not let it accumulate near the floater. The bulk flows generated in water during particle propulsion also disperse the ethanol released from the hydrogel. The ethanol concentration profile in this cylinder (with uniform initial concentration and boundary condition of zero concentration at its one open end), as a function of time, is given by,

\[ C = \frac{4C_0}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)^2} e^{-D(2n+1)^2 \pi^2 t / 4l^2} \cos \left( \frac{(2n+1)\pi x}{2l} \right) \]

where \( C_0 \) is the initial concentration of ethanol in the gel (7.9 \( \times \) 10^{-2} g/cm\(^3\) for 10% ethanol by volume and 39.5 \( \times \) 10^{-2} g/cm\(^3\) for 50% ethanol by volume), \( l \) is the length of the gel body (4 mm) excluding the PDMS plug, \( d \) is the inner diameter of the particle (1 mm), \( D \) is the ethanol-water counter-diffusion coefficient (1.1 \( \times \) 10^{-9} m\(^2\)/s for 10% ethanol by volume and 0.5 \( \times \) 10^{-9} m\(^2\)/s for 50% ethanol by volume), and \( x \) is the distance from the insulated circular end of the cylinder.

We integrated this concentration profile over the length of the cylinder to obtain the total amount of ethanol \( Q \) contained in the hydrogel at any time \( t \), given by,

\[ Q = \frac{2C_0 d^2}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n}{(2n+1)^2} e^{-D(2n+1)^2 \pi^2 t / 4l^2} \sin \left( \frac{(2n+1)\pi l}{2l} \right) \]
We use the ethanol-water counter-diffusion coefficient because as ethanol is released from the hydrogel, water diffuses into the gel because of osmosis (hydrogel has higher affinity for water than for ethanol).\textsuperscript{3} Counter-diffusion of water also decreases the ethanol concentration in the gel with time. Using eqn. 2, the net amount of ethanol released from the gel-based floater in between two consecutive pulses, $\Delta Q$, can be obtained by,

$$\Delta Q = Q(t_n) - Q(t_{n+1})$$

where $t_n$ is the time at which the $n^{th}$ pulse occurs. We analyzed the digital movies of the experiment on a frame-by-frame basis to obtain the intervals $t_1$, $t_2$, $t_3$, ..., $t_n$, $t_{n+1}$ at which the particle pulsates, and by using eqn. 3 calculated the $\Delta Q$ for each pulse. We plotted $\Delta Q$ versus time for gel boats with 10% and 50% ethanol by volume in the hydrogel, and obtained that ~0.3 $\mu$g (the $\Delta Q$ value at which the plots terminate, indicating negligible particle motion) is the minimum amount of ethanol needed by these gel boats to propel in water by overcoming the viscous drag (Figure S1).

**Scheme S1.** Schematic illustrating the parameters in the ethanol transport model. The floater was modeled as a cylinder, having uniform initial ethanol concentration $C_0$, with one end open to mass-transfer. The bulk concentration of ethanol $C_b$ was assumed to be negligible (zero) at the open end, based on the propulsion mechanism. $\Delta Q =$ ethanol released from the hydrogel for a pulse, $l =$ length of the particle, $d =$ inner diameter of the particle, $\gamma =$ surface tension.
Plot of $\Delta Q$ values for gel boats

We plotted $\Delta Q$ versus time for gel boats with 10% and 50% ethanol by volume in the hydrogel, and obtained that ~0.3 $\mu$g (the $\Delta Q$ value at which the plots terminate, indicating negligible particle motion) is the minimum amount of ethanol needed by these gel boats to propel in water by overcoming the viscous drag. The procedure used for obtaining these $\Delta Q$ values has been described in the previous section.

Figure S1. Plots of $\Delta Q$ versus time for gel boats with (a) 10 % and (b) 50 % initial ethanol concentration (by volume) in the gel.
Figure S2. Experimental data of the (a) pulse interval ($\Delta t$) and (b) total distance ($d'$) propelled in each pulse, fitted to eqns. 5 and 8, respectively, for floaters with 50 % vol. initial ethanol concentration in the gel. The obtained values of the fitting parameters have also been shown.

The obtained values of the fitting parameters $k_1$ and $k_2$ are different for floaters with 10% and 50% ethanol concentration in the hydrogel (Fig. 4 in paper, Fig. S2). The higher magnitude of $k_1$ for the case of 10% ethanol in the hydrogel is because it will take more time for a given $\Delta Q$ amount of ethanol (released from the hydrogel) to accumulate on the surface in this case relative to 50% ethanol in the hydrogel. Similarly, higher magnitude of $k_2$ for the case of 50% ethanol in the hydrogel is obtained because for a given $\Delta Q$ amount of ethanol released from the hydrogel, the distance propelled by the particle will be larger relative to 10% ethanol in the hydrogel because of more rapid rate of ethanol transport to the surface.
Six movies for visualizing the motion of the particles and the propulsion mechanism are provided (in Windows WMV format)

- **Pulsating_motion.wmv (Movie 1)**

  The movie shows the pulsating motion of the gel boat in water - the cycle of sudden motion followed by rest. The ethanol concentration in the polyacrylamide hydrogel is 10% by volume of the liquid. The movie is speeded up to 4×, and is recorded with Sony DSC-V1 Cyber-Shot digital camera.

- **Control_experiment_propulsion_mechanism.wmv (Movie 2)**

  The movie displays the propulsion mechanism of the gel-based particle. Red colored food dye is mixed with ethanol to trace its release profile from the particle placed at the bottom of the Petri dish. A cycle of surface flows followed by its disruption can be clearly observed. The particle remains stationary, being at the bottom of the Petri dish. The movie is speeded up to 2×, and is taken with an Olympus SZ-61 optical microscope focused on the side of the Petri dish.

- **Back_forth_pulsating_motion.wmv (Movie 3)**

  The movie illustrates the back and forth pulsating motion of a gel floater open to mass-transfer at both ends of the hydrogel. The ethanol concentration in the polyacrylamide hydrogel is 10% by volume of the liquid. The movie is speeded up to 4×, and is recorded with Sony DSC-V1 Cyber-Shot digital camera.

- **Rotation angled-edge.wmv (Movie 4)**

  The movie shows the periodic rotational motion of a gel-boat with an angled edge. The ethanol concentration in the polyacrylamide hydrogel is 50% by volume of the liquid. The movie is speeded up to 2×.

- **Partially closed end_tango.wmv (Movie 5)**

  The movie illustrates the asymmetric back and forth motion of a gel-boat with an open end and a partially closed end. The ethanol concentration in the polyacrylamide hydrogel is 50% by volume of the liquid. The movie is speeded up to 4×.

- **L-shape.wmv (Movie 6)**

  The movie displays the motion of a floater comprising of two sub-particles joined together into an L-shape. The ethanol concentration in the polyacrylamide hydrogel is 50% by volume of the liquid. The movie is speeded up to 4×.
Details of the derivation of eqn. 7 in paper

Assuming additivity of surface tension and constant molarity of the solution,

\[ \gamma = x_e \gamma_e + (1-x_e) \gamma_w \]

\[
\frac{d\gamma}{dC} = \frac{d\gamma}{d\left(\frac{x_e N}{V}\right)} = \frac{V}{N} \frac{d\gamma}{dx_e} = \frac{N}{V} (\gamma_e - \gamma_w) = \text{constant}
\]

where \( \gamma \) = solution surface tension, \( \gamma_e \) = ethanol surface tension, \( \gamma_w \) = water surface tension, \( x_e \) = ethanol mole fraction, \( C \) = ethanol concentration, \( N \) = total number of moles, \( V \) = total volume of solution.

Data for the motion of a two-way dancing particle

Table S1. Pulse interval and distance propelled corresponding to the pulses originating from the two ends of a particle consisting of a 50% ethanol hydrogel at one end and a 10% ethanol hydrogel at the other. The results obtained from the model are in reasonable agreement with the experiments.

<table>
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<th>Expt. (50%)</th>
<th>Model (50%)</th>
<th>Expt. (10%)</th>
<th>Model (10%)</th>
<th>Expt. Ratio (50%:10%)</th>
<th>Predicted Ratio (50%:10%)</th>
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<td>0.09</td>
<td>0.09</td>
<td>2.16</td>
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