The mechanics of hydrogel crawlers in confined environment

Franck Vernerey and Tong Shen

Mechanical Engineering, University of Colorado Boulder, 427 UCB, Boulder, CO, USA

Cite this article: Vernerey F, Shen T. 2017 The mechanics of hydrogel crawlers in confined environment. *J. R. Soc. Interface* 14: 20170242.

We present theoretical and experimental results regarding the development of temperature-sensitive hydrogel particles that can display self-motility in confined channels. Inspired by the motility of living organisms such as larva, the motion of the particle relies on the combination of two key mechanisms. The first, referred to as actuation, is enabled by the cyclic extension and retraction of the particle owing to oscillations of its temperature around the so-called lower critical solution temperature. The second, referred to as symmetry breaking, transforms the isotropic particle actuation into a directed motion owing to the asymmetric friction properties of the channel’s surface. The role of particle confinement in these processes is, however, less intuitive and displays an optimal value at which the particle’s step size is maximum. These observations are supported by a model that identifies the underlying locomotion mechanisms and predicts the dependency of the particle motion efficiency on the confinement condition, as well as frictional properties of the substrate. Our analysis suggests that the existence of a lubrication layer around the particle hinders its motion at low confinement, while an excessive degree of confinement is detrimental to the particle’s overall deformation and, thus, to its locomotion efficiency.

1. Introduction

In the past decades, the dynamical behaviours of soft active particles and swarms have captured the interest of the soft matter community. Indeed, these systems including ant colonies [1], flocks of birds [2] and schools of fish [3] have shown to possess intriguing behaviours and applications to robotic swarms [4,5]. From a medical point of view, a potentially interesting system is that of migratory particles that can move through the pore space of a material in order to perform functions such as delivering drugs, restoring tissue functions [6,7] or destroying unwanted tissues including tumours [8]. In contrast with passive particles (liposomes and vesicles for instance) [8] and micro-swimmers [9–12], such particles are required to move through confined spaces using contact forces. Very little is known about physical processes that can achieve this goal. Fortunately, locomotion in confined spaces is common in nature, providing many possibilities for bioinspired particles. Indeed, organisms such as cells, bacteria, larva and worms [13,14] often migrate through porous spaces using adhesion or frictional forces to propel themselves forwards.

In this paper, we find inspiration in the locomotion mechanism of a simple, yet ubiquitous class of organisms that consists of larva and maggots. Maggots and larval diptera typically crawl by using telescoping peristaltic motion [13], which relies on a periodic elongation and shortening of their body. While this mechanism alone would not produce motion, the dermis of larvae is covered with sharp ratchet-like microspikes [13,15] (figure 1b) which effectively entitles it with asymmetric frictional properties. During the peristaltic cycle, this feature breaks the symmetry in frictional forces between the front and the back of the organism and leads to directed motion. The velocity of the larva was then found to be proportional to its stride length $\Delta s$ and stride frequency $1/t_c$ in the form [13]

$$V = \frac{\Delta s}{t_c} \quad (1.1)$$
Compared to limbed invertebrates, observed velocities remain low, but due to their soft bodies, larva and maggots may possess a distinct advantage in moving through confined spaces and through narrow pores [16]. Reproducing the locomotion mechanisms in a synthetic system therefore requires the use of an active material that undergoes periodic deformations and an asymmetric surface. Polymeric materials, such as hydrogels, liquid crystal elastomers [17,18] and shape-memory polymers are excellent candidates, due to their ability to undergo large deformations under external stimulus or the presence of internal chemical reactions. In this context, Yang et al. [19] used electrically triggered periodic bending of a hydrogel sheet to achieve sustained ‘walking’ on a ratched surface while Maeda et al. [20] incorporated a chemical oscillator, known as the Belousov–Zhabotinsky (BZ) reaction, to periodically deform the hydrogel sheet. Motion in confined space was further pursued by generating a BZ-induced peristaltic wave along a hydrogel body in order to mimic the body contraction of earthworms [21,22]. This approach, however, did not produce enough deformation to enable sustained motion. Yeghiazarian et al. [23] therefore proposed to use heat, instead of a chemical reaction, to locally trigger hydrogel deformation and produce larger peristaltic deformations. Although this latter approach enabled particle locomotion in a confined capillary tube, this solution remains limited because manually localized stimulation cannot be used for remote systems.

In this study, we develop a bioinspired particle that migrates in a straight confined channel using a combination of periodic deformation, remotely triggered by periodic heating and cooling, and the asymmetric friction produced by the structure of its surroundings. Because the stimulus is not locally applied on the particle, this strategy has the potential to enable remote actuation via magnetic heating [24] and the locomotion of particle swarms in a porous medium. Here, we concentrate on a single particle for which the locomotion mechanisms are rationalized by the development of a theoretical model. We particularly focus on how the particle deformation, friction and confinement affect the size of its stride, and eventually its velocity through equation (1.1). While this work concentrates on the motion in a straight channel, it will provide a stepping stone towards the development of active particles that can invade porous media in particular biological tissues.

2. Experimental set-up

In this section, we first describe the design of a hydrogel cylindrical crawler and its environment, which consists of a narrow, cylindrical channel coated with surface patterns. We discuss the fabrication of the system, as well as the experimental analysis of the particle motion, together with key results.

2.1. Fabrication

To mimic the oscillating peristaltic deformation of larva, we fabricated cylindrical thermo-sensitive Poly(N-isopropylacrylamide) (PNIPAAm) particles whose fully swollen diameter and length were \( R_0 = 0.8 \) cm and \( \ell = 3 \) cm, respectively. PNIPAAm is a heavily studied temperature-responsive polymer [25,26] that displays a reversible phase transition at a lower critical solution temperature (LCST) between 32°C and 36°C. Below the LCST, the hydrogel is hydrophilic and absorbs large amounts of water to achieve its swollen state. Above the LCST, the hydrogel becomes hydrophobic and dehydrates, losing almost 90% of its volume. This material therefore presents an ideal candidate for the synthesis of self-actuated particles under periodic temperature oscillation around the LCST. The hydrogel particle was then confined in a hollow cylindrical channel and was reversely immersed back and forth into a cool fluid \( (T = 22 \, \text{C}) \) and a hot fluid \( (T = 42 \, \text{C}) \) in order to enable a periodic deformation. To prevent the gel from sticking to the channel walls [27], we formed an intercalating surfactant layer by immersing the particles in 0.4 wt% Tween 20 solution until the gel was fully swollen. For observation purposes, the particles were also dyed red by storing them in 2 wt% Ferroin solution. In nature, the existence of confinement provides the essential three-dimensional environment for organisms to protrude and achieve sufficient friction/adhesion for motion. For instance, Jacobelli et al. [28] reported that there was a degree of confinement for which the amoeboid locomotion of T cells was optimized. To explore the effect of confinement with our system, we therefore fabricated channels with different radii \( R = 0.4, 0.35, 0.3 \) and 0.25 cm, all smaller than the initial particle radius \( R_0 \) for experiment. A measure of particle confinement could thus be defined as follows:

\[
c = \frac{R_0}{R} - 1.
\]
2.2. Active motion

As described above, once confined in the channel, the particle was subjected to a periodic oscillation in temperature around \( T_c \) in order to obtain a series of extension–contraction cycles. The timing of these cycles was finely adjusted such that the particle reached its maximum swollen length during the extension stage and its original unswollen length during the contraction stage. We note that these values were different depending on the particle’s degree of confinement as discussed in more detail in appendix A. On average, the expansion time was on the order of 2 min, while the retraction stage was slower, on the order of 6 min. Particle motion was measured during this process by measuring the position of its geometric centre for different surface features and confinements. A total of four measurements were made per cycle, for a total number of eight swelling–de-swelling cycles (figure 3a). With these results, the particle’s average step size was estimated by performing a linear least square fitting of the particle’s translation as a function of the number of cycles. For isotropic

Figure 2. (a)(i) Schematic of experiment set-up; the dyed hydrogel particle is placed in the three-dimensional printed channel with anisotropic surface. During the experiment, the channel is immersed in the water bath. (ii) Photograph of the hydrogel particle next to the three-dimensional printed cylindrical channel. (b) Schematic plots showing the periodic change of temperature of the fluid and the resultant swelling/de-swelling of the hydrogel. (c) Sequential snapshots of a hydrogel in one de-swelling/swelling cycle in a channel with anisotropic patterns. The step size \( \Delta s \) of motion can be measured as the effective displacement of the particle centre. Scale bar, 1 cm.

With this definition, \( c = 0 \) when the unswollen particle has the same radius \( R \) as the channel, while confinement increases with decrease in channel radius \( R \). For particles with a radius of \( R_0 = 0.5 \) cm, the confinement measure becomes \( c = 0.25, 0.43, 0.67 \) and 1 for the printed channels described above.

In biological systems, this asymmetry generally results from structural asymmetries on the organism’s surface, as observed in the belly scale structure of snakes [29,30] and maggots [15] for instance. Reproducing such features on a soft deformable particle is challenging, but a simpler alternative is to directly coat the channel with a ratchet-like structure whose asymmetry can be well defined. To introduce this asymmetry, the internal surface of the channels was therefore printed with periodic ridges of spacing \( w = 3 \) mm and height \( h = 0.8 \) mm. The asymmetry was induced by laterally moving the tip of the ridges by a distance \( s \) as shown in figure 2a as characterized by the non-dimensional parameter \( s/w \). We see here that \( s/w = 0 \) corresponds to symmetric ridges, while \( s/w = 0.5 \) indicates extreme asymmetric ridges. For each confinement condition \( c \), four channels with ratchet patterns \( s/w = 0, 0.2, 0.3 \) and 0.5 were fabricated using T-glass material due to its transparency and thermostability. All the channels were printed by the Lulzbot 3D printers that possess sub-millimetre resolution.

Figure 3. (a) Motion of particle centre in the channels for a confinement \( c = 0.67 \) and ratchet patterns \( s/w = 0, 0.3 \) and 0.5 in eight subsequent cycles. It is shown that the particle achieves greater effective displacement when the ratchet pattern is more anisotropic. (b) Experiment results of the motion of particles in the channels with different ratchet patterns and confinement conditions. It is shown that anisotropic patterns are favourable for motion and there exists an optimal confinement \( c \) for largest step size \( \Delta s \). (Online version in colour.)
ratchets \((s/w = 0)\), the particle exhibits a small and random motion with a zero effective step size. A clear directional motion was however observed as \(s/w\) increases to a step size \(\Delta s = 0.32 \text{ cm}\) at \(s/w = 0.5\) and a fixed confinement \(c = 0.67\).

The role of confinement was also explored by performing the active motion test for the four degrees of confinement described above \((c = 0.25, 0.43, 0.67\) and \(1)\). Results regarding the particle step size as a function of ratchet asymmetry and confinement levels are shown in figure 3b. While the trends show a strong correlation between the level of asymmetry and the particle’s step size, for all \(c\), confinement plays a more intricate role. Indeed, our results suggest that both small and large confinements tend to hinder motion and that there exists an optimal confinement, around \(c = 0.5\), for which we observe the maximal step size.

3. Mechanics of active sliding in confinement

To rationalize the above observations and further investigate how the particle velocity depends on frictional properties \([31,32]\), confinement \([28,33,34]\) and geometry \([35]\), we next construct a mathematical model of the physical processes leading to particle locomotion (figure 4).

3.1. Model

The model considers a cylindrical particle confined in a channel of radius \(R\) (figure 4a) that, under a change in temperature, swells to a new, stretched state as shown in figure 4b. The swelling process is monitored here by a decrease in the polymer volume fraction \(\phi\) from its original value \(\phi = \phi_0\) to its hydrophilic value \(\phi = \phi_0/\phi\). When the channel surface is asymmetric, this deformation generates a translation of the particle that can be studied by balancing forces acting on a particle segment (figure 4c). For this, we first note that experimentally, the particle deformation occurs at a time scale of minutes, while inertial forces would require accelerations on the order of seconds to be comparable to frictional and swelling forces. For this reason, we neglect initial forces and only consider the equilibrium of the internal stress \(\sigma_i\) and the frictional shear stress \(\tau\) resulting from the particle–channel interactions. This yields the force balance equation:

\[
R\sigma_{ix} + 2\tau = 0, \tag{3.1}
\]

with boundary conditions \(\sigma_i(R \pm L/2) = 0\) describing the stress-free conditions at the particle’s extremities. In the following, we use the symbol \(X\) to denote the position of a material point in its reference position and a symbol \(x = x(X)\) for its position in the swollen state. We particularly seek to express the particle deformation field and stresses as functions of the fixed reference state \(X\). We further note here that the dynamics are controlled by gel swelling, a phenomenon that happens at a time scale that is much slower than the sliding process. It is, therefore, reasonable to assume that friction is a quasi-static, equilibrium process, well described by dry friction. The shear stress \(\tau\) is therefore related to the magnitude of the normal pressure \(\sigma_n\) on the particle through an asymmetric Coulomb’s friction model \([32]\):

\[
\begin{align*}
\tau(X) &= -\mu^-\sigma_n(X) \quad \text{if} \ u(X) < 0 \\
\tau(X) &= -\mu^+\sigma_n(X) \quad \text{if} \ u(X) > 0,
\end{align*}
\tag{3.2}
\]

where \(u\) is the axial displacement of a material point on the particle, while \(\mu^-\) and \(\mu^+\) are the friction coefficients associated with forward and backward sliding, respectively. To estimate the stresses in the particle during the swelling process, we built a model based on the Flory–Rehner theory \([36]\) whose details are shown in appendix A. Our analysis shows that the longitudinal stress \(\sigma_x\) can be expressed as follows:

\[
\sigma_x = G \left( 1 + \frac{1}{3} c^{1/3} \frac{\phi_{1/3}}{\phi} - \phi \right) + \frac{k_B T}{v} \ln(1 - \phi) + \phi + \chi \phi, \tag{3.3}
\]

where \(c\) was defined in equation (2.1), \(G\) is the shear modulus of the gel in its dry state, \(\chi\) is the Flory–Huggins parameter, \(k_B T\) is the thermal energy and \(v\) is the specific volume of solvent. The first term in (3.3) corresponds to the elastic stress, while the second is the swelling force (or osmotic pressure). In a nutshell, when the Flory–Huggins parameter suddenly changes from its hydrophobic values \(\chi_0\) to its hydrophilic value \(\chi_s\) due to cooling, the hydrogel particle sees an increase in its osmotic pressure that produces the extension necessary to ‘step forward’. Again, a thorough discussion of the corresponding derivation and associated physical processes is provided in appendix A. Similarly, the normal stress \(\sigma_n\) generated by the channel confinement could be calculated as follows:

\[
\sigma_n = \frac{G d \phi^{1/3}}{3} \left( \frac{\phi_{1/3}}{\phi} - \phi \right). \tag{3.4}
\]

Here, we used \(\phi_0 = \phi_0(1 + c)^3\). This function shows that the normal stress vanishes in the reference state if \(c = 0\), while it monotonically increases with swelling (or decreasing \(\phi\)). Substituting the above stresses into governing equations (3.1) and (3.2), we obtain the following nonlinear equation for the polymer fraction \(\phi(X)\):

\[
\begin{align*}
&\frac{k_B T}{G v} \left( 1 + 2 \chi(\phi - 1) - \left( \frac{\phi_{1/3}^2}{\phi} - 1 \right) \right) \frac{\partial \phi}{\partial X}
\end{align*}
\]

\[
- \frac{2 \mu^+}{3R} \left( \frac{\phi_{1/3}^2}{\phi} - 1 \right) = 0, \tag{3.5}
\]
where the coefficients $\mu^\pm$ depend on position. The derivation for equation (3.5) can be found in appendix B. In the back of the particle (figure 5b), over a length $L^-$, the velocity $u^-$($X$) is negative and the friction coefficient is $\mu^-$. In the front of the particle, over a length $L^+$, the velocity $u^+$($X$) is positive and the friction coefficient is $\mu^+$. The boundary between these two domains has a zero velocity and is therefore called the anchor point (AP) in the remainder of this paper. Its location $X$ can be determined by ensuring the continuity of stress $\sigma_x$ together with the stress-free boundary condition at the particle’s end ($\sigma_x(L^\pm) = 0$). This can be done by directly integrating equation (3.1) on $[0, L^\pm]$ and each side of the domain in order to obtain

$$\alpha = \frac{(L^+ - L^-)}{L} = \frac{(1 - \mu^+ / \mu^-)}{(1 + \mu^+ / \mu^-)}.$$

(3.6)

The non-dimensional parameter $\alpha$ designates the relative position of the AP in the particle. If $\alpha = 0$, the AP is at the centre of the particle, while if $\alpha = 1$ or $\alpha = -1$, the AP is located at the back or front edges of the particle, respectively. Equation (3.6) indicates that the position of the AP is influenced by both friction asymmetry and confinement; for symmetric friction ($\mu^+/\mu^- = 1$), or low confining pressure ($c \rightarrow 0$), the AP is located at the particle centre ($\alpha = 0$), while for the case $r = \mu^+/\mu^- < 1$, the AP moves to the back of the particle during the swelling stage and to its front during de-swelling (figure 5b). A solution for $\phi(X)$ can further be obtained numerically on both sides of the AP by integrating equation (3.5) for constant coefficients on fixed domains. In this paper, we use a nonlinear, finite-difference solver and plot the results in terms of the strain field $\epsilon(X)$, measured from the particle’s reference, unswollen state ($\phi = \phi_0$) as follows:

$$\epsilon(X) = \lambda(X) - 1 = \frac{\phi_0}{\phi(X)} - 1,$$

(3.7)

where $\lambda(X)$ is the longitudinal stretch ratio between the unswollen and swollen states. Figure 5 shows how the location of the AP and the strain field $\epsilon$ along the particle are influenced by the friction coefficient $\mu^+$, ratio $r = \mu^+ / \mu^-$ and the degree of confinement $c$. Overall, the strain field varies non-linearly along the particle with its lowest value at the AP and maximum value at the particle’s extremities. For small friction ($\mu^- < 1$) or low confinement ($c < 0.1$), strains increase and become more homogeneous along the particle. For very large friction and confinement, the deformation is mostly concentrated at the particle’s end. Finally, as expected, we see a shift of the AP towards the back of the particle as the ratio $r = \mu^+ / \mu^-$ increases.

### 3.2. Particle step size

To better understand how particle strains affect its motion, we now introduce a simplified analysis based on the above results. For this, the swelling–de-swelling cycle is assumed to occur in a symmetric fashion, such that the overall step size $\Delta s$ can be expressed in terms of the location $\alpha$ of the AP which shifts from the back of the particle to its front between the swelling and de-swelling stage (figure 6a). Defining $\bar{\epsilon}$ as the average strain in the effective domain of the particle between the two APs, with length $\alpha L_0$ in the reference state, we show in figure 6b that the translation of the particle centre during one cycle (or step size) is expressed by

$$\Delta s = \alpha L_0 \bar{\epsilon},$$

(3.8)

where $L_0$ is the reference length of the particle. We see here that there are three ways for a particle to increase its step size: (i) increase the distance $\alpha$ of the AP from the particle’s centre via substrate anisotropy according to equation (3.6); (ii) increase its unswollen, reference length $L_0$, which is a direct function of the dry particle length $L_d$ and its level of confinement $c$. Indeed, squeezing a particle in a narrow channel effectively increases its length according to the following relation:

$$L_0(c) = \frac{L_d R}{R_d}(1 + c)^{3/2}.$$

(3.9)

Finally, (iii) the model predicts that step size increases with average strain $\bar{\epsilon}$ between unswollen and swollen

---

**Figure 5.** (a) Solution of the normalized strain $\epsilon/\epsilon_s$ along the particle with different values of $\mu^+$, $r$ and $c$, where $\epsilon_s$ is the strain of the particle in its confined state when friction vanishes. We could see that $r$ affects the anisotropy of particle deformation, while both $\mu^+$ and $c$ have large effect on the strain along the particle. (b) The motion of the particle in one swelling/de-swelling cycle. The colour on the particle surface indicates the displacement; the black ring shows the position of the AP, while the red ring depicts the position of the particle centre.
4. Particle motion in ratcheted channels

The above model predicts the particle step size as a function of both confinement and the channel’s frictional properties. To relate these findings to surface morphology, this section first aims to establish a relationship between friction, confinement and ratchet design. These findings are then used to make predictions of the particle motion in different environments. Comparisons with experimental observations are provided. 

4.1. Role of surface patterns and confinement on friction

To determine how friction depends on surface patterns $s/w$ and particle confinement $c$, we thus sought to experimentally measure the coefficients $\mu^+$ and $\mu^-$ in various conditions by evaluating the critical pressure $\Delta P_\text{c}$ at which particle motion stalled in both the forward and backward directions, respectively. For a given normal pressure (determined from equation (4.1)), we estimated the shear stress on the particle surface at the onset of motion by $\tau = R \Delta P_\text{c} / 2L$, where $L$ is the particle’s length.

Results depicted in figure 7 show that, for larger confinements, the shear stress increases linearly with normal stress as predicted by the Coulomb friction. It is further observed that the difference between forward ($\mu^+$) and backward ($\mu^-$) friction coefficients increases linearly with substrate anisotropy $s/w$, which allows us to write

$$\mu^\pm = \mu_\infty + b \frac{g}{s/w},$$

where $\mu_\infty$ is the friction coefficient for symmetric patterns and $b$ measures the sensitivity of friction to substrate patterns. A table of the measured parameters is provided in appendix C. When the degree of confinement is reduced below 0.5, we observe that the above results deviate from the Coulomb friction, a phenomenon that can be explained by the existence of a lubrication layer between the particle and the channel [37]. Indeed, for small confining pressure, friction is dominated by viscous effects within this layer and is insensitive to surface patterns. Experiments show that the friction coefficients exhibit an exponential decay as the degree of confinement is reduced to zero (figure 7c).
which is well captured by the following function:

\[ \mu^\pm = \mu_0^\pm + (\bar{\mu} - \mu_0^\pm) \exp \left( \frac{-c}{\epsilon_0^\pm} \right), \quad (4.2) \]

where \( \epsilon_0^\pm \) is the measure of the critical confinement at which the system transitions from viscous to dry friction [37] and \( \bar{\mu} \) is the low dynamic friction coefficient at zero confinement.

### 4.2. Predictions of particle motion in different environments

To confirm whether the model presented in §3 can explain particle locomotion, we now compare the experimental results obtained in figure 3 solely based on equations (3.6) and (3.8)–(3.10) and the relationship between the friction coefficient and the ratchet design shown in equation (4.1). To replicate the experimental conditions in the model, we consider a particle of dimensions \( L_0 = 0.8 \) cm and \( R_d = 0.28 \) cm in its dry state, which was swollen to an unconstrained radius \( R_0 = 0.5 \) cm before encapsulation into the channel. The different confinement conditions \( c = 0.25, 0.43, 0.67 \) and 1 were discussed in §2. In these conditions, figure 8b depicts how the variables \( \alpha, L_0 \) and \( \epsilon_{AP} \) depend on confinement for a high substrate asymmetry \( s/w = 0.5 \) as predicted by the model. We generally see that confinement favours a shift of the AP towards the particle’s extremities and an effective increase in the particle size \( L_0 \) both of which promote a larger step size. However, these trends are counterbalanced by the fact that particle strain exponentially decreases with confinement. Overall, the model predicts that the step size originally increases with confinement.

---

**Figure 7.** (a) Photo of ratchet patterns with different anisotropy \( s/w \). (b) Experimental measurements of frictional shear stress \( \tau \) between a hydrogel particle and channels with different ratchet patterns \( s/w \) under different normal stresses \( \sigma_n \) which was controlled by the confinement \( c \). (c,d) The calculated friction coefficient \( \mu \) from (b) for different ratchet patterns \( s/w \) and confinement conditions \( c \). The lines in (c) are fits of the \( \mu - s/w \) relationship using equation (4.1) and the lines in (d) are fits of the \( \mu - c \) relationship using equation (4.2). (Online version in colour.)

**Figure 8.** (a) Graphical explanation of how the confinement condition \( c \) affects the values of variables \( \alpha, L_0, \epsilon \) and \( \Delta s \). In this example, \( s/w = 0.5 \). Three schematic particles in swollen states are shown, where the notation \( AP_s \) denotes the anchor point for swelling and \( AP_u \) denotes the anchor point for de-swelling. (b) Experimental measurement of particle motion under different values of \( c \) and \( s/w \) are shown by symbols, while the model predictions are shown by lines. For each ratchet pattern \( s/w \), the optimal confinement and step size are depicted by the green line. For these studies, the length of the particle in its dry state is \( L_d = 0.8 \) cm. (Online version in colour.)
until it reaches a maximum around $c = 0.5$ and then starts decreasing. We see that for low confinement, the particle is able to undergo large deformation, but its motion is hindered by the weak asymmetry in the location of the AP. By contrast, large confinements favour asymmetry extension but hinder the overall particle deformation. An optimal step size is found as a compromise between these two situations. To complete our study, we then compared model predictions to experimental measurements for different values $s/\omega$ of ratchet designs and confinement $c$. Results, reported in figure 8b, show that within this range of parameters, the model accurately describes the particle step size. We see here that large ratchet asymmetry is a requisite for particle motion, while confinement should be appropriately tuned to maximize the motion of the crawling hydrogel particle. The closed form expression for the step size in terms of system design can further be used to optimize our system. In that context, by setting $\partial\Delta s/\partial c = 0$, figure 8b shows the level of confinement that is necessary to enable a maximum step size for different values of friction asymmetry $s/\omega$. We observe here that optimum confinement decreases nonlinearly with substrate asymmetry.

## 5. Conclusion

In summary, we have developed a soft self-crawling hydrogel particle that can propel itself in a confined channel owing to a combination of cyclic swelling–de-swelling and the asymmetric friction properties of the surrounding environment. The mechanisms behind motion were identified and understood by a model that can accurately predict how particle locomotion depends on the surface patterns and confinement conditions. From experimental and modelling studies, we have particularly shown that motion is affected by three main attributes: particle length, frictional asymmetry with its substrate and the degree of confinement. A closed-form relationship between the particle step size and these attributes was constructed based on a combination of analytical and numerical modelling. Overall, the model shows that step size scales linearly with particle length and increases nonlinearly with the substrate asymmetry, measured by the ratio $\mu^-/\mu^+$. The role of confinement is, however, subtle because it may or may not favour particle motion. Low confinements result in a weak normal force and a switch to the hydrodynamic lubrication regime between the particle and the channel, which results in a drastic reduction in the friction asymmetry and step size. By contrast, high confinement favours asymmetry, but significantly hinders the particle deformation for motion. We found that, for our system, an optimum confinement around $0.2 \leq c \leq 0.5$ was therefore required for optimal step size, depending on the friction properties. We note that the model only considered the swelling phases of particle motion, assuming that the de-swelling behaves similarly. Good agreements between model prediction and experimental measurements suggest that this assumption was satisfactory to capture the motion mechanism. Furthermore, while step size is an important aspect of locomotion, the rate (or characteristic time $t_c$ from equation (1.1)) at which the particle expands and retracts similarly plays a key role in motion efficiency. While it is expected that this time decreases with decreasing particle size and increasing hydrogel permeability, the specific solvent flux boundary conditions on the particle–channel interface will probably play a role. A thorough study of this process is left for future studies.

From an application standpoint, our experimental system may be thought of as a first generation of crawling particles in confined environments that can be actuated remotely. Although motion was studied in a one-dimensional channel, it provides useful information on how to achieve efficient motion for the more general problem of crawling through tortuous paths. In this case, the ratchet structure will need to be transferred to the surface of the soft matter, mimicking the patterns seen on a variety of maggot species [38] and other scaled organisms [39–41]. Although such an approach will call for more sophisticated numerical techniques [42,43], it is expected that the role of confinement and surface asymmetry uncovered in this study will still hold for these more complex systems.

### Data accessibility

All data from experiment and model are presented in the paper.

### Authors' contributions

F.V. conceived and designed the study and wrote the manuscript. F.V. and T.S. carried out the analytical derivations. T.S. carried out all other experiments and simulations. All authors read and corrected earlier versions of the manuscript and approved the final version.

### Competing interests

We declare we have no competing interests.

### Funding

We acknowledge the support from the National Science Foundation (DMR-1411320) and the National Science Foundation under the NSF MRSEC (DMR-1420736).

### Acknowledgements

We thank Marti Garriga Font for his help with channel preparation, experimental set-up and the discussions about the model. We thank Anung Golla for his help with the experimental set-up. We thank Mark Stoykovich for his suggestions on hydrogel fabrication and lubrication.

### Appendix A. Oscillating hydrogel in a confined channel

The theory describes hydrogel swelling through its free energy $\Delta G = \Delta G^{\text{el}} + \Delta G^{\text{mix}}/\phi$, where $\phi$ is the volume fraction of the polymer. This functional represents the competition between the polymer–solvent mixing force, described by the free energy of mixing $\Delta G^{\text{mix}}$ and the elasticity of the cross-linked polymer, described by the elastic free energy $\Delta G^{\text{el}}$. The mixing force is captured by the free energy:

$$\Delta G^{\text{mix}} = \frac{k_B T}{\rho} \left[ (1 - \phi) \ln(1 - \phi) + \chi \phi(1 - \phi) \right],$$  \hspace{1cm} (A 1)

with $k_B$ being the Boltzmann constant, $T$ the temperature and $\rho$ the specific volume of solvent. The polymer–solvent interaction parameter $\chi$ abruptly changes from its room temperature value of $\chi = 0.243$ to $\chi = -0.818$ above the critical temperature $T_c = 32^\circ C$ [44]. This phenomenon is responsible for the unswellling of the particle above $T_c$, as discussed in the previous section. Following standard rubber elasticity [45], the elastic contribution is captured by a neo-Hookean stored elastic energy function of the form

$$\Delta G^{\text{el}} = \frac{G}{2} \left[ \text{tr}(F^T F) - 3 - 2 \ln(\det(F)) \right],$$  \hspace{1cm} (A 2)

where $F$ is the deformation gradient that maps the deformation of the polymer from its dried state and $G = \rho_k k_B T$ is the shear modulus of the gel in its dry state (with $\rho_k$ being the number of polymer chains per unit volume). Using an unconfined compression test on cylindrical particles of PNIPAm used in our
Figure 9. (a) Particle in its unswollen equilibrium state, unswollen non-equilibrium state (reference in our experiments) and swollen equilibrium state. (b) Elastic longitudinal stress and osmotic pressures \( \sigma_1 = \pi(x_1) \) and \( \sigma_2 = \pi(x_2) \) in terms of polymer fraction \( \phi \). Equilibrium points A and C are found when the stresses are equal to the osmotic pressures. (c) Geometry parameters of the channel and the particle, and the free body diagram that shows the longitudinal stress \( \sigma_l(x) \) and the frictional shear stress \( \tau \) acting on the particle. (Online version in colour.)

experiments and a calibration method described in [46], \( G \) was estimated to be 158 \( \pm \) 15 kPa.

To evaluate these energies for our system, we consider a cylindrical particle with length \( L_0 \) and radius \( R_0 \) in its dry state. The particle is then brought to its unswollen, reference state with radius \( R_0 \), length \( L_0 = L_0 R_0 / R_d \) and polymer volume fraction \( \phi_0 = (R_d / R_0)^3 \). As it swells in a channel of radius \( R = R_0 / (c + 1) \) (equation (2.1)), its deformation history can theoretically be split into an unconstrained swelling stage (the particle is not in contact with the channel) and a constrained swelling stage. During the free swelling stage, the deformation gradient is isotropic with non-zero components \( F_x = F_r = \phi^{1/3} \), where the subscripts \( x \) and \( r \) indicate the longitudinal and radial directions, respectively. Under these conditions, we see that the particle radius equals the channel radius \( R \) when the polymer fraction reaches a critical value \( \phi_c = (R_d / R_0)^3 \), or in terms of confinement

\[
\phi_c = \phi_0 (1 + c)^3. \tag{A 3}
\]

In our study, we are particularly interested in the constrained swelling stage during which the particle’s radial extension is blocked by the channel and the deformation gradient becomes \( F_x = \phi^{1/3} / \phi \) and \( F_r = \phi^{1/3} \). Using these results, the Cauchy stress tensor \( \sigma \) in the particle can be obtained from the Gibbs free energy using the relation

\[
\sigma = l^{-1}(\partial \Delta G / \partial F)^T \quad \text{where} \quad l = \det(F).
\]

The deformation gradient \( F \) that maps the deformation of the polymer from its dried state can be written as

\[
F = \text{diag}(\lambda_{x1}, \lambda_{x2}, \lambda_d), \tag{A 4}
\]

where \( \lambda_{x1}, \lambda_{x2} \) and \( \lambda_d \) are the elongations of the particle along the three principle directions in the cylindrical coordinates. From equation (A 4), the change of volume of the particle \( V = \det(F) = \lambda_{x1} \lambda_{x2} \lambda_d \), the volume fraction of polymer can be obtained as \( \phi = 1 / (\lambda_{x1} \lambda_{x2} \lambda_d) \). We recall that the total free energy is written as \( \Delta G = \Delta G^{\text{el}} + \Delta G^{\text{mix}} / \phi \) where the free energy of mixing \( \Delta G^{\text{mix}} \) and the elasticity \( \Delta G^{\text{el}} \) were shown in equations (A 1) and (A 2), respectively. The Cauchy stress \( \sigma \) in the particle can be obtained from the free energy by

\[
\sigma_1 = f^{-1} P_i F_{x3} = \frac{G}{2} \left( \frac{\lambda_d}{\lambda_{x1} \lambda_{x2} \lambda_d} - \frac{1}{\lambda_{x1} \lambda_{x2} \lambda_d} \right) - \pi \tag{A 5}
\]

\[
\sigma_l = f^{-1} P_i F_{r1} = \frac{G}{2} \left( \frac{\lambda_d}{\lambda_{x1} \lambda_{x2} \lambda_d} - \frac{1}{\lambda_{x1} \lambda_{x2} \lambda_d} \right) - \pi
\]

where \( \pi \) is the osmotic pressure:

\[
\pi(x, \lambda) = -\frac{k_B T}{v} \ln(1 - \phi) + \phi + \chi \phi.
\]

In the free swelling regime, \( \lambda_{x1} = \lambda_{x2} = \lambda_d = \lambda \) and \( \phi = 1 / \lambda^3 \). In this regime, the elastic stresses can be written as

\[
\sigma_1^e = \sigma_l^e = G \left( \frac{1}{\lambda} - \frac{1}{\lambda^3} \right) = G \phi (\phi^{-2/3} - 1). \tag{A 6}
\]

In the constrained swelling regime, \( \lambda_{x1} = \lambda_{x2} = R_d / R_0 \). In this way, we can write \( \phi_c = (R_d / R_0)^3 \) and \( \phi = \phi_c^{1/3} \lambda_d \) and the elastic stresses can then be written as

\[
\sigma_1^e = G \left( \frac{\phi_{c}^{1/3}}{\phi} - \phi \right) \quad \text{and} \quad \sigma_l^e = G \phi (\phi_{c}^{-2/3} - 1). \tag{A 7}
\]

When the Flory–Huggins parameter suddenly changes from its hydrophobic value \( x_0 \) to its hydrophilic value \( x_S \) due to cooling, the hydrogel particle sees its internal osmotic pressure jump from \( \pi_0 = \pi(x_0) \) to \( \pi_S = \pi(x_S) \) as shown in figure 9. When \( \pi = \pi_0 \), the osmotic pressure is low and the particle is unswollen, but still in contact with the channel because \( \phi_0 < \phi_c \). When \( \pi = \pi_S \), the particle inflates.
significantly ($\phi_0 < \phi_n$), producing the extension necessary to ‘step forward’. In the absence of friction, the stretch ratio $\lambda$ between the unswellen and swollen states can indeed be calculated as $\lambda(X) = \phi_0 / \phi_n$, which is approximately equal to 2 for our system. The normal pressure between the hydrogel and channel $\sigma_n$ is finally obtained as the difference between the radial elastic stress $\sigma_r'$ and the hydrostatic pressure $p$, as follows:

$$\sigma_n = \sigma_r' - p = \frac{Gd^2/3}{3} \left( \frac{\phi_c - \phi}{\phi_c} \right),$$

(A 8)

### Appendix B. Governing equation for particle motion

At equilibrium, the internal stress $\sigma_r$ is balanced by the frictional shear stress $\tau$:

$$R\sigma_{r,x} + 2\tau = 0,$$

with the stress-free boundary conditions at the particle’s end $\sigma_r(\pm L^x) = 0$. As it was discussed, the shear stress $\tau$ is related to the magnitude of normal pressure $\sigma_n$ through the asymmetric Coulomb’s friction model as $\tau^\pm = -\mu^\pm \sigma_n$. The above equation is solved in the reference configuration, for which equation (B 1) can be rewritten as

$$R\sigma_{r,x} + 2\frac{\phi_c}{\phi} \frac{\partial \phi}{\partial x} = 0.$$

In the above equation, $\sigma_{r,x}$ can be obtained from equations (A 5) and (A 7) as

$$\sigma_{r,x} = \left[ -G \left( \frac{\phi_c^{-2/3}}{(\phi_c)^2 + 1} + \frac{k_h T \phi}{v \left( 1 + 2\chi (\phi - 1) \right) \phi^2} \right) \frac{\partial \phi}{\partial x} \right].$$

(B 3)

We further substitute the normal stress $\sigma_n$ appearing in equation (A 8), and finally obtain the nonlinear equation for polymer volume fraction $\phi(X)$ shown in equation (3.5):

$$\frac{k_h T \phi}{Gv \left( 1 + 2\chi (\phi - 1) \right)} \left( \frac{\phi_c^{-2/3}}{(\phi_c)^2 + 1} + \frac{2\mu^\pm}{3K} \left( \frac{\phi_c}{\phi} \right)^2 - 1 \right) \frac{\partial \phi}{\partial X} = 0.$$

(B 4)

The numerical solution for $\phi(X)$ can be obtained by directly integrating the above equation on $[0, L^x]$ at each side of the domain.

### Appendix C. Model parameters

See table 1

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Meaning</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G$</td>
<td>shear modulus of hydrogel</td>
<td>158 kPa</td>
</tr>
<tr>
<td>$V$</td>
<td>specific volume of water</td>
<td>1 cm$^3$ kg$^{-1}$</td>
</tr>
<tr>
<td>$R_d$</td>
<td>radius of hydrogel at dry state</td>
<td>0.28 cm</td>
</tr>
<tr>
<td>$L_d$</td>
<td>length of hydrogel at dry state</td>
<td>0.8 cm</td>
</tr>
<tr>
<td>$\chi$</td>
<td>polymer–solvent interaction parameter</td>
<td>0.243</td>
</tr>
<tr>
<td>$b$</td>
<td>The constant parameter in equation (4.1)</td>
<td>1.453</td>
</tr>
<tr>
<td>$c^0$</td>
<td>critical confinement for viscous–dry friction transition</td>
<td>0.1748</td>
</tr>
<tr>
<td>$\bar{\varepsilon}$</td>
<td>the constant parameter in equation (3.11)</td>
<td>0.9</td>
</tr>
</tbody>
</table>

### References