Autonomous lablet locomotion and active docking by sensomotory electroosmotic drive

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Abstract

In this paper, we show how autonomous microscopic CMOS particles, called lablets, here at the scale of 100µm, are equipped with an electroosmotic drive, and calculate that this can propel them sufficiently to overcome the strong viscous drag at this length scale, depending on the ionic strength and pH of the solution in which they are immersed. We then use this thrust and the currently fabricated lablet design to show in simulation how programmable movement of lablets can be obtained. The lablets are equipped with chemical sensors and simple programmable functionality that we show can form a substrate for chemotactic behaviour and autonomous docking to a target surface (such as a second lablet). We calculate the concentration field induced by an electrically active lablet and show that other lablets can dock by chemotaxis in this field. Lablets can self-assemble to form compartments, and perform chemical operations, and hence provide a potential electronic-chemical substrate for building artificial cells. The investigation of autonomous motion capabilities is an important basic functionality for particles of this size, where passive Brownian motion is already slow, when motion has been implicated as a fundamental property of early life [1].

Introduction

Electrochemical and electrophoretic locomotion of passive microparticles in solution has recently attracted significant attention, with bimetal rods presenting impressive unswitched locomotion speeds [2], but would need to be combined with extremely miniaturized directional control and intelligent sensing and switching functions to support full robotic potential at this micrometer scale. In a major initiative, our group together with European partners is building smart electrochemical autonomous agents at the scale of 100µm using specially post-processed CMOS chips that we call lablets [3], since they can dock together to form tiny autonomous labs for doing chemistry in a high surface to volume ratio environment.

Previously, we have shown the power of microfluidic-technology incorporating electroosmotic flow [4] (EOF) in addition to electrophoresis in concentrating DNA as well as controlling flows [5,6]. In this paper, we show how autonomous microscopic lablets that we have designed are equipped with an electroosmotic drive, and calculate that this can propel them sufficiently to overcome the strong viscous drag at this length scale, depending on the ionic strength of the solution in which they are immersed. In this simulation, we first calculate the magnitude of the expected electroosmotic thrust for lablets, taking pH effects into account, and then use this thrust in the current lablet design to show how programmable movement of lablets can be obtained. The lablets are equipped with simple programmable functionality, in the first instance this can be described by timed finite state machines, but full microcontroller functionality is achievable if the electronics resolution is enhanced from the current scale of 180nm down to 65nm, using special transistor designs that involve slow switching and low leakage currents being developed by an electronics partner [7].

The lablets are equipped with several sensors that allow pH to be measured in an internal channel at two different points, which also allows gradients to be detected for close proximity docking. However, for chemotactic tracking, gradients are too weak for direct detection using closely spaced sensors, and instead sequential sensor measurements at different positions of a spiraling trajectory show effective chemotaxis of lablets. The paper shows how such motion can be generated with simple two state control of actuation, and explores the extension from lablet chemotaxis to docking, which can allow pairs of lablets to form specific compartments and communicate chemical and electronic information. The paper concludes with a discussion of the implication of lablet docking for living technology processes and a lablet life cycle.

Lablet architecture and design

The basic design of the lablets involves an internal branched channel with the topology of a T and with bent channel ends to permit rotational thrust from electro-osmotically dispelled fluid. A typical lablet is shown in fig. 1. Essentially each lablet consists of a 100x100µm area of a thinned CMOS wafer (40-50 µm), so that two face-to-face lablets can form a cube, with an active enclosed channel network along the mid axis. Alternatively, a single lablet can be fabricated with a laminating film (10-20µm) to enclose the channels. The detailed lablet electronic design and fabrication will be described elsewhere. The lablets are fabricated in standard CMOS (180nm process, Europractice via IMEC/TSMC) in collaboration with AIS [7] and T.Maeke at the 20cm wafer scale. Post processing of the lablets includes structured metal recoating of the electrodes (PVD, lift-off, Al to Au), surface

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topography changes (using the photoresist SU8), specific galvanic post processing of sensor electrodes (IrO2) and supercap inter-digitat structures (MnO2), and lamination. Subsequently, wafers are thinned to 40μm and diced on a supporting substrate to produce the array of labelts. The laminate needs to be photo-structured, also to allow its removal in the saw-lines between labelts to prevent tearing during wafer sawing. The supercap is required to store sufficient charge (from a wireless charging cycle) to support 1000s of electrolyte double layer potential switches. Typical capacitances of 20x20 μm electrodes in solution are 50-100 pF (depending on the electrolyte) whereas the supercap (enclosed on the other side of the labelts) can achieve μF capacitances.

Figure 1. Labelt architecture. **Left**: Schematic of labelt T channel (bottom) and 3D assembly of a pair of labelts showing enclosed channel exits (light blue-green color). **Right**: Light microscope image of 100x100μm CMOS labelt substrate, with metal electrodes showing white. Overlaid labels show the three actors A0-2 and the two interior sensors S0-1 with reference electrode SR, in the T shaped channel. The channel is bordered by raised insulating segments (1μm grey) and closed from above either with a laminate or a second docked labelt. The power and docking electrodes (labeled in grey, V0-1,D0-1) are not needed during locomotion.

Labelt electronics is currently equipped with a programmable finite state machine (programmable bit-serially by means of daisy chained flip-flops), and have been made in a combinatorial set of variants, which will be published separately. For the purposes of this article, the following functionality is required:

(i) to be chargeable and run autonomously for a period of 20 min.
(ii) to sense a chemical signal (e.g. pH) and detect if it exceeds a threshold
(iii) to activate either one of two electrode configurations, (a) A0 + A1 (b) A0 + A2, for a fixed duration of time, depending on sensor signal from (ii)
(iv) to be programmed to support a periodically recurring sequence of activity involving simple sequences of actions of type (ii) and (iii).

**Electroosmotic thrust calculation**

The use of electroosmotic forces to drive fluid through a channel is well known in microfluidics, and was employed by us to fill reaction chambers and filter DNA [5]. The theory of electroosmotic drive is well developed [8,9] so we simply record the main equations used in the standard model for the current work. In this formulation, we consider the ion system to be in equilibrium, so it is primarily governed by Poisson Boltzmann equation. Using these assumptions, we can calculate the electroosmotic thrust produced by coupling with the simplified Navier Stokes equation. In the simplest case where the Debye layer is thin compared with the height of the channel, the electroosmotic velocity is given by

$$ v = -\frac{\varepsilon(\zeta_1 + \zeta_2)}{\eta} E_z $$

(1)

where the velocity is proportional to the applied field $E_z$ along the channel (in z direction), to the sum of the zeta-potentials of the top and bottom surfaces, and to $\frac{\varepsilon E_z}{\eta}$ i.e. the ratio of dielectric constant to dynamic viscosity of the solution. Typical mobility values of $5 \times 10^{-8}$ m²/Vs give $v = 1$ mm/s for 2V over a 100μm length channel.

At low ionic strengths, a fuller analysis involves the velocity at any height $y$ in the channel of height $h$, which can be derived from the force exerted on charges along the channel by means of Poisson’s equation

$$ F_z = E_z \sigma = E_z \varepsilon \frac{\partial^2 \psi}{\partial y^2} = \eta \frac{d^2 v}{dy^2} $$

(2)

where $\psi$ is the local electrostatic potential, and $\sigma$ is the local charge density. The final equality in (2) equates this force to the viscous shear force from the Navier Stokes equation.

Labelt surface channels are predominantly SiO₂ coated and we employ the analysis of [10] for such surfaces[11] in to calculate the zeta potential. The charge density for a partially deprotonated SiO₂/SiOH labelt surface is $\sigma = -\varepsilon \Gamma_{SiOH}$ where $\Gamma = \Gamma_{SiOH} + \Gamma_{SiOH}$ is the total surface density, and

$$ \frac{[H^+]_0 \Gamma_{SiOH}}{\Gamma_{SiOH}} = 10^{-pK_a}, \quad [H^+]_0 = 10^{-pH} $$

(3)

where the acid concentration near the surface $[H^+]_0$ is related to its value in the bulk, $[H^+]_0 = [H^+]_0 \exp(-\psi_{d}/k_B T)$, by the surface potential $\psi_{d}$. Furthermore, the potential drop across the Stern layer with capacitance $C = \sigma/(\psi_{d} - \psi_{a})$ relates this surface potential to the zeta potential $\zeta \equiv \psi_{a}$ or potential at the inner edge of the diffuse double layer. The six equations in this paragraph can be reduced to a single equation relating $\sigma$ to $\psi_{d}$, eliminating $\Gamma \Gamma_{SiOH}, \psi_{0}$ in favor of known quantities $\Gamma, pH, pK_a, C$. From equilibrium Poisson Boltzmann theory, [12], $\psi_{d}$ is the potential at the inner edge of the diffuse layer and given by

$$ \sigma \left( \psi_{d} \right) = \frac{2e \phi_{d} e}{\beta e} \sinh \left( \frac{\beta e \psi_{d}}{2} \right) $$

(4)

and so these two equations for \( \sigma, \psi_d \) can be solved to find \( \zeta \equiv \psi_d \). The potential at any height \( y \) in the channel of height \( h \) can then be found from

\[
\psi(y) = \frac{4kT}{e} \left[ \tanh^{-1} \left( \tanh \left( \frac{e\zeta}{4kT} \exp \left( -\kappa y \right) \right) \right) \right] + \left( \tanh^{-1} \left( \tanh \left( \frac{e\zeta}{4kT} \exp \left( -\kappa(h - y) \right) \right) \right) \right]
\]

(5)


Figure 2. Lablet drift speed under electroosmotic drive. The drift speed for a 100x100\( \mu \)m lablet (hydrodynamic friction set equivalent to a 50\( \mu \)m radius sphere) is calculated as a function of pH (red points) for a concentration of 10mM monovalent salt (e.g. NaCl) and for pH 8 as a function of salt concentration (blue points). Higher drift velocities of several lablet diameters per second can be obtained using a combination of high pH (e.g. 10) and high concentration salt (100mM). Logarithms are base 10. Calculations were performed with the following values of constants, cf.: pH=10, \( C=2.9 \) F/m\(^2\), \( pK_a = 7.5 \), \( f = 8/\text{nm}^2\), bulk ionic concentration 10 mM, applied voltage 2 V, height of channel 1\( \mu \)m.

The solution of eq.(3) with slip length \( b \), typically 20nm, is then

\[
v(y) = -\frac{e\zeta}{n_0} \left[ \left( 1 - \psi(y) \right) - \frac{b}{\zeta} \left( \frac{c}{n_0} \right) \right] E_z
\]

(6)

and the induced thrust on the lablet can be computed from the mean velocity \( \bar{v} \) over the channel as

\[
F_T = \rho w h \left( \frac{eE_z}{n_0} \right)^2 \left( \frac{1}{h} \int \left( 1 - \psi(y) \right) dy - \frac{b}{\zeta} \left( \frac{c}{n_0} \right) \right)^2
\]

(7)

where the solution has density \( \rho \) and \( w \) is the width of the channel (height \( h \)). The linear and rotational drift velocities of lablets (assuming cube = sphere) are

\[
\nu = \frac{F_T}{6\pi n_0 r} \quad \omega = \frac{F_T \sin \alpha r}{8n_0 r^3}
\]

(8)

where the angular velocity in the second equation is equal to the torque on the lablet induced by the thrust from the base of the T in fig. 1 (with fin angle typical value \( \alpha = 30^\circ \)).

The energy efficiency of the electroosmotic drive can be calculated from the current consumption during operation. There are two contributions to the current: the conductive current and the convective current densities \[9\]

\[
j_{\text{cond}} = 2e \frac{c e E_z}{n_0} \left( 1 - \frac{c}{n_0} \right) - b \left( -\frac{\sigma}{\zeta} \right)
\]

(9)

\[
j_{\text{cond}} = 2e \frac{c e E_z}{n_0} \left( \frac{c}{n_0} \right) - b \left( -\frac{\sigma}{\zeta} \right)
\]

(10)

where both expressions need to be integrated over the channel, \( c \) is the bulk ion concentration as in fig. 2 and \( \mu \) is the salt average ion electrophoretic charge mobility, corresponding to typical room temperature ion diffusion coefficients of \( 10^{-3} \) m\(^2\)/Vs, i.e. \( 3.9 \times 10^{-8} \) m\(^2\)/Vs. The resulting values of the currents at pH 10 and 10mM salt concentration are 39 nA and 5.6 nA. Lablet supercaps can only support powers up to 2nA for 1000s without recharging. An additional factor of 20 is needed for continuous mobile operation at low power densities. Note that the thrust is proportional to the velocity squared while the current consumption is proportional to the velocity, so that this factor can be regained at higher fluid expulsion velocities. Using concentrations of 100mM instead of 10mM at pH 11 increases lablet velocity by a factor of 4 to 5, and current efficiency by over a factor of 2. A factor of roughly 10 in efficiency enhancement is still needed to run locomotion continuously for 20 minutes. This can be achieved using a nozzle structure at the end of channel. For example, reducing the 20\( \mu \)m channel width to a 2 \( \mu \)m opening for 1 \( \mu \)m length will only increase the overall hydrodynamic resistance of the propulsion channel by a factor of 14\%, decreasing the interior channel velocity by this amount, while increasing the expulsion velocity and hence the current efficiency by a factor of 8.5. This enables us to reach the locomotion target of navigation for over \( 1/4 \) hr. Since lablet actuation can be operated for a fraction of the time, resulting in slower motions, efficiency can be regained for our original scenario, and we prefer to continue to work with the slower lablet velocities calculated above, ignoring the nozzle enhancements in the following.

**Programmable locomotion of lablets**

We first consider the deterministic motion of a lablet under thrust in 2D. Translational and rotational Brownian motion add jitter to this movement and allow a lablet trajectory to explore space more fully. As we saw in Section 2, for 100\( \mu \)m lablets this is indeed a perturbation. Motion in 3D can be achieved through interaction with buoyancy control (e.g., using electro-induced gas formation via electrolysis) see below and discussion/conclusions section.
Figure 3. Elementary programmable motions of labllets in 2D. The 8 distinct period-5 two state lablet actuation patterns (see text) are used to generate these trajectories. The duration of the elementary lablet translation and rotation thrusts was same and equal to 3 sec.

Perhaps the simplest form of programmable motion involves switching between just two possible thrust modes, straight and bent, modes 1&2, as described in section 2, involving switching between two possible actuation states. A finite memory of 4 bits for example can then encode a sequence of 4 such binary states, resulting in the 16 elementary motions on cyclic repeat. However not all are different when repeated in a continuous stream, and in fact there are only 6 different trajectories 0000, 0001, 0011, 0101, 0111, and 1111. Similarly, for 5 bits, the 32 elementary motions reduce to eight distinct trajectories 00000, 00001, 00011, 00101, 00111, and 11111. We show the trajectories created by these eight standard motions in fig. 3. Each cycle of steps involves a constant net rotation and translation, so that indefinite iteration fills out a circling spiral trajectory provided not all steps are pure translation. Note that depending on the duration of the elementary thrust steps, these deterministic trajectories can approximate space-filling curves. With either longer period sequences or by including a larger number of elementary modes, or by switching between two or more different periodic patterns more complex and divers trajectories can be obtained. Three such different patterns with different resulting curvatures, as shown in fig. 3R are used in the following section.

Figure 4. Periodic trajectories using 1, 2 and 3 elementary rotations interleaved with translation, which are the basic operations used in the next section for chemotaxis. The steps in the deterministic trajectories are integrated analytically in Mathematica both for the pure translation step (mode 1) and the combined translation step (mode 2) and then combined into trajectories.

The equations describing the trajectories of the elementary segments are thus
\[
x'[t] = \begin{cases} 2v \cos[\theta[t]] & \text{mode}[t] = 1 \\ v(\cos[\theta[t]](1+\sin[a(x)]) - \cos[a(x)]\sin[\theta[t]]) & \text{mode}[t] = 2,3 \end{cases}
\]
\[
y'[t] = \begin{cases} 2v \sin[\theta[t]] & \text{mode}[t] = 1 \\ v(\cos[a(x)]\cos[\theta[t]] + (1+\sin[a(x)])\sin[\theta[t]]) & \text{mode}[t] = 2,3 \end{cases}
\]
\[
\theta'[t] = \begin{cases} 0 & \text{mode}[t] = 1 \\ \alpha & \text{mode}[t] = 2 \end{cases}
\]

Where mode 1,2 and 3 involve electrodes \{A0,A1\}, \{A0,A2\} and \{A1,A2\} respectively. If the voltages are reversed the signs of all time derivatives also reverses (the lablets move backwards). The magnitude of Brownian fluctuations perturbing these trajectories is shown in the examples of the following section.

The combination of these trajectories with buoyancy control can lead to complex motion in 3D. Although coupling between buoyancy induced motion and lateral motion is possible via hydrodynamics (e.g. disk settling instability) we do not consider such effects in the current work.

**Lablet concentration sensing and locomotion**

As with mobile single celled organisms, mobility coupled with concentration sensing can allow the organism to navigate to find food in an aqueous environment. Different mechanisms of such chemotaxis are known, including the tumbling mechanism in bacterial chemotaxis [13] and the spiral drift of sperm chemotaxis [14]. Since rotational diffusion is slow on the 50µm radius scale of lablets, compared with driven rotational speeds of ca. 0.6 rad/s, the random tumbling of bacteria can be replaced by deterministic directed rotation. Inspired by the elegant chemotactic mechanism for sperm cells [14], which has been modeled as a continuous propagation with curvature \(K(t)\) governed by

\[
K(t) = c_1 + c_2 \frac{dc}{dt}
\]

where \(c\) is the chemotactant concentration and \(c_1\) and \(c_2\) are constants, we can attempt to capture part of such a mechanism with a lablet sensor-actuator program.

A very simple program that should be able to perform similar chemotactic lablet locomotion is of the form

```plaintext
repeat
1. sensor_value_old = S0-SR // read sensor value in interior channel
2. activate A0+A1 for 10 clock cycles // activate translation drive
3. sensor_value_new = S0-SR // read new sensor value
4. activate A0+A2 for 1 clock cycles // activate rotation drive
5. if (sensor_value_new>sensor_value_old) then
   activate A0+A2 for 1 clock cycles // alternate rotation drive
until end
```

We employed a number of variants of this simple program to investigate lablet chemotactic capabilities for several variants of this algorithm discussed below. We used the steady state solution to the diffusion equation either is spherical or in cylindrical coordinates to define the concentration field. The solution could be expressed analytically with either 1/r or

log(r) dependence. In cylindrical coordinates, employed in fig. 5, two boundary conditions were used: concentration \( c = 100 \) units at \( r = 0.1 \) \( \mu m \), \( c = 0.01 \) at \( r = 4.2 mm \). The time for each thrust operation was set to 0.4 secs. The tablets were programmed to sense concentration values after four thrust operations.

Since current tablets are only equipped with a binary threshold (analog-digital conversion) on read, with 2-4 levels, the signals read in steps 1 and 2 are 0-1 or 0-3. In fig. 5 we compare absolute threshold sensing, with very limited bit resolution with continuous sensing, including the effects of sensing noise. Limited sensing resolution restricts chemotactic capabilities when absolute thresholding is employed. A dynamic scaling of sensitivity in analog-digital conversion will be required for robust chemotaxis. If the concentration surrounding tablets are far from the defined threshold, they will continue spanning the region, programmed by the thrust operations.

![Figure 5. Trajectories of tablets exhibiting chemotaxis.](image)

Due to harmonic nature of these equations, they are conformally invariant. For the limiting case of fast reactions (essentially at equilibrium), the boundary conditions also becomes conformally invariant. So, by mapping the analytical solution for one dimensional case, we could create concentration field around two coplanar electrodes. We used Schwarz–Christoffel mapping [16] to map one dimensional solution for concentration and potential into two parallel electrodes to coplanar electrodes. The one dimensional solution for concentration, potential [15] and expression for mapping are given by:

\[
\nabla^2 c = 0, \quad \nabla \cdot (c \nabla \phi) = 0
\]

expression for mapping are given by,

\[
c = 1 + J \text{Im}(f(t))
\]

\[
\phi = \log \left( \frac{1 + J \text{Im}(f(t))}{K(1 - J)^2} \right), \quad J = \tanh \left( \frac{V}{4} \right)
\]

\[
f(t) = \int \left( \frac{K}{(t - t_1)^{3/2}(t - t_2)^{3/2}(t - t_3)^{1/2}(t - t_4)^{1/2}} \right) d(t)
\]

where, $J$ is the current density scaled to limiting current density, $k$ is a constant determined by matching. So, we used following parameters to create two dimensional profile of concentration field, applied voltage 0.5V, bulk concentration 10mM, characteristic length scale of 10mm, dimensionless rate constant as 0.1. We interpolated the concentration field after mapping the coplanar electrode geometry from the unstructured grid shown in Fig 6A. We defining the initial position of lablet, we sensed the chemical field and perform locomotion similar to the scheme described in the last section. The plotted trajectories in Fig. 6c shows, successful docking of the lablet on the electrode. We observed in the certain limit of chemical noise, lablet, in all the cases, docks on the electrode independent of the starting position of the lablet.

Figure 6. A) Conformal map created by mapping parallel plate electrodes geometry to coplanar electrodes using Schwarz-Christoffel mapping, B) Concentration sensed by lablet over time (starting position shown in C, red point in vicinity of grey curve). The plots show that with increase in the noise in chemical sensing the total time taken to dock increases. C) Trajectory of the lablet from two different starting locations (shown by red points) until it reaches the chemotractant emitting electrode (white, e.g. low pH source).

Work is currently in progress on using various combinations of the forward and reverse modes 1-3 in equation (11) to extend this translational docking of lablets to a lablet emitter chemotractant signal to a full rotational and translational docking model and will be reported at the conference. In further work, we are also exploring the interaction of electroosmotic drive with electrochemical buoyancy control. An initial estimate, indicates that currents of 1nA can create gas bubbles which change the buoyancy of a lablet by 25pN in 10 seconds, which should be sufficient for vertical locomotion with buoyancy compensated lablets. Buoyancy compensated lablets have also been studied experimentally by F. Stepanek using attached light oil droplets (personal communication).

**Conclusions: Implications for artificial life**

Whereas, much emphasis is placed on the integration of chemical reactions for artificial wet life, and electronic chemical lablets appear an attractive platform to construct working artificial life systems (assuming that the unprogrammed CMOS lablet is regarded simply as a substrate consumable or building block), currently becoming accessible scales of ca. 100µm for smart active chemical particles require special mechanisms of motion to allow lablets to interact. Interaction of lablets with lablet substrate is essential to a lablet reproduction life cycle, in which a programmed active lablet can dock with a new substrate lablet (unprogrammed) and modify its chemical coating or interior chemicals to allow transmission of program information, prior to undocking release.

Lablet pairing is a novel form of dynamical compartmentation control that can allow lablets to operate on shared chemicals without dilution or interference from the bulk solution. Locomotion is necessary to support pairing, and while this could potentially be achieved by external perturbations (e.g. stirring), it is certainly more efficient to consider autonomous locomoted docking procedures as studied in this paper. In another contribution [17], partners in the MICREAgent project [3], have investigated swarming behavior of lablets assuming an abstract model of sensing and control of motion. The current work complements this study with a more detailed physical investigation of lablet locomotion potential and docking. Whereas much attention has been attracted by the self-propulsion capabilities of oil droplets [1] and bipolar microrod particles [18,19], the current paper shows how programmable control of electrodes can be combined on smart particles at somewhat larger scales to ensure directed locomotion.

In conclusion, lablets with active electronic programs able to interact with chemistry provide a novel substrate for artificial life research. This paper demonstrates that the same mechanisms of electrode actuation and sensing suitable for the control of chemical reactions on lablets can be harnessed to provide autonomously navigating lablets under electroosmotic drive. Careful engineering of channel geometries (e.g. nozzles) will be necessary to ensure sufficient efficiency in thrust for prolonged operation, but the paper establishes that rather simple control mechanisms can then be used to perform...
complex tasks such as chemotaxis and docking. Docking of lablets allows a novel control of dynamic chemical compartmentation as a substrate for an artificial lablet life cycle as discussed above. Further work in this direction is in progress.

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