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Wireless powering of e-swimmers

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Miniaturized structures that can move in a controlled way in solution and integrate various functionalities are attracting considerable attention due to the potential applications in fields ranging from autonomous micromotors to roving sensors. Here we introduce a concept which allows, depending on their specific design, the controlled directional motion of objects in water, combined with electronic functionalities such as the emission of light, sensing, signal conversion, treatment and transmission. The approach is based on electric field-induced polarization, which triggers different chemical reactions at the surface of the object and thereby its propulsion. This results in a localized electric current that can power in a wireless way electronic devices in water, leading to a new class of electronic swimmers (e-swimmers).

The design of structures capable of moving under the influence of external stimuli enables new applications ranging from roving intelligent micro-sensors to integrated systems for drug delivery. To this extent, biological motors and swimming micro-organisms inspire the development of artificial micro- and nano-machines^{1–5}. The most popular developed strategy utilizes a chemical fuel such as hydrogen peroxide to induce motion: a catalytically active patch⁶ on a Janus particle breaks the symmetry of the system and induces motion⁷. Both self-electrophoresis and bubble propulsion have been explored in this context⁸. Recently, new techniques for writing microstructures on surfaces⁹, transporting cargo¹⁰, delivering cargo in microfluidic channels^{11,12}, and sensing¹³ have been reported. Different groups are concentrating their efforts on the design^{14,15}, the synthesis^{16–18} and the motion control of chemical swimmers to push forward this interdisciplinary field of research. Because the use of hydrogen peroxide as a chemical fuel is not compatible with biological applications, alternative motion mechanisms are also attracting considerable attention, such as the use of magnetic field induced propulsion, which is very efficient at the microscale^{17,19–21}, or light driven motion allowing to envision microrobotic systems^{22,23}. It is also possible to harness an electric field^{24,25} to induce motion at the macro-²⁶ and the microscale²⁷. In this general context of mobile microscale robotics²⁸ we show in the present contribution that it is possible to combine motion with the localized generation of electric power to operate electronic devices in water and thus develop integrated micro-rovers. In this case, an electric field polarizes a conducting object and gives rise to water splitting at its surface, which locally generates bubbles that propel the object. We demonstrate that at the same time the electric current flowing inside the object can be used to drive electronic devices, such as light-emitting diodes (LED).

A polarization voltage ΔV is induced across the object in solution, which is proportional to the electric field E and to its characteristic dimension l ²⁹.

$$\Delta V = E l \quad (1)$$

If this polarization voltage is high enough, the opposite sides of the object experience different potentials that can give rise to spatially separated electrochemical reactions, namely an oxidation at the anodic pole and a reduction reaction at the cathodic pole. Attractive applications of such bipolar electrochemical processes have been developed in the fields of analytical chemistry³⁰, materials science^{31–33} and also for the propulsion of objects^{10,34,35}.

A major challenge in this context is to integrate functionalities into the moving objects. Therefore we propose here to use the local electric current that flows inside a bipolar electrode not only to achieve motion control, but also to power electronic devices, such as an integrated temperature sensor or LEDs. Velev *et al.* have successfully demonstrated that LEDs can be powered in solution by AC fields and perform motion²⁵ based on a local electroosmotic ion flux. The approach proposed here relies on bubble-induced propulsion of conducting objects that can be achieved with bipolar electrochemistry due to water splitting at the opposite extremities of a bipolar object, according to the following reactions:



This intrinsic electrochemically-induced asymmetry can be exploited to promote directional motion in aqueous solutions (Figure 1). At the same time, electrons flow across the bipolar electrode from the oxidation-side (reaction (3) at the δ^+ side) to the side where protons are reduced (reaction (2) at the δ^- side). This local current can power, as we show here, an electronic device, which enables integrated *e*-swimmers. The AC powering²⁵ has the advantage of eliminating electroosmosis and electrophoretic effects, however the advantage of the DC powering is that through the controlled release of bubbles steering is easier. In an AC device the diodes can only move in one direction, whereas in the DC set-up reported here, reversal of the direction is possible, which makes the two approaches very complementary.

The involved redox reactions could potentially generate local changes in pH around the swimmer³², however as the swimmer is moving, and thus creating convection, these domains of different pH immediately remix to result in the initial pH of the solution.

Results

Contactless power-transfer to electronic devices in solution. In a first step we will demonstrate that different electronic devices can be wirelessly powered in an aqueous solution under the influence of an external electric field. The induced polarization leads to an electron flow from the δ^+ side of the object to the δ^- end. When an electronic element is incorporated in the path of electron flow it is possible to operate the device without physical contact to the feeder electrodes. The first example, illustrated in Figure 2A, is the operation of a light-emitting diode. Three independent diodes are placed in water and exposed to the electric field established between the anode and the cathode of the electrochemical cell. As can be seen from the side view in Figure 2B, the polarization leads to the reduction of protons on the right connector of the LED, whereas oxygen evolution is observed on the left side. Due to the intrinsic stoichiometry of water electrolysis, the amount of generated hydrogen is twice as high as that of oxygen, and this is reflected by the respective quantities of visible rising gas. The amplitude of the concomitant light emission can be tuned by adjusting the external electric field. However there is a threshold

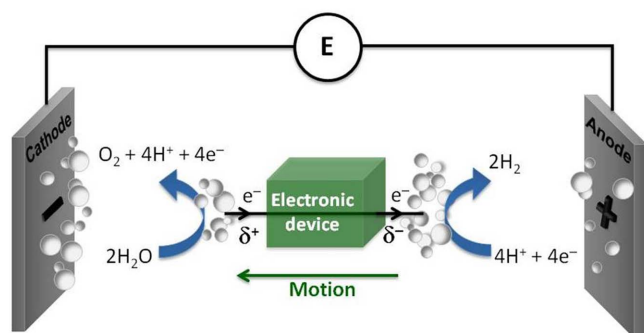


Figure 1 | Schematic illustration of the *e*-swimmer concept. The input and output (black arrows) of an electronic device are immersed in an aqueous solution and exposed to an external electric field (applied by the gray feeder electrodes). The resulting polarization (δ^+ and δ^-) leads to an oxidation and a reduction reaction on the opposite sides of the object (blue arrows), triggering an electric current through the object. This current is used to power an integrated electronic device and the simultaneously developing gas bubbles are causing the object's motion in the solution. The corresponding cell design is illustrated in Figure S1.

to overcome which is the difference in formal potential of the two involved redox reactions. Once the polarization voltage is higher than this value, there is a classic exponential Butler-Volmer dependence between the overpotential experienced by both sides of the bipolar object and the reaction rate (current) on these two sides. This means that the higher the polarization voltage the higher the current/bubble production and therefore also the light emission.

Besides LEDs, more sophisticated electronic circuits can also be powered based on this concept, as can be seen in Fig. 2C. It illustrates the operation of a complex electronic device capable of performing multiple tasks in a contactless way and in an aqueous environment. An electronic sensor powered by the bipolar current measures the local temperature and provides an analog signal to an embedded central processing unit. After signal treatment, a numerical signal is generated and transmitted from an embedded infrared emitter to a receiver located outside of the water reservoir. This signal can be converted into a measure of the temperature (Figure 2D), thus allowing to track for example the ohmic heating of the solution (for details see SI). These proof-of-principle experiments demonstrate that bipolar electrochemistry is able to deliver a stable power input for useful electronic devices. Technologies required to miniaturize such kind of devices are available³⁶, allowing to design systems that run with currents in the μA range and/or with a power as low as tens of nW at 0.5 V. Thus one can imagine driving already relatively sophisticated circuits without causing significant Joule heating, because the currents through the solution will then be more than thousand times lower. In this way multiple functionalities could potentially be integrated, leading to remote controlled *e*-swimmers.

Propulsion of *e*-swimmers. In this section we show how multiple electric fields can be used to impart directional motion to *e*-swimmers. We recently reported levitation experiments with glassy carbon beads as swimmers¹⁰, but they did not allow for motion in more than one direction. In the present case we propose a concept for the dynamic control of the motion in several directions, independently of the cell geometry, and to simultaneously integrate an electronic functionality.

A rocket-like swimmer, composed of an outer plastic shell (b), combined with two metal wires (a) that act as bipolar electrodes (Figure 3A, 3B), can be moved by controlling the kinetics of the gas bubble production. The tip of the rocket has a tapered opening through which gas bubbles can escape. If the rate of the bubble formation exceeds the release rate, gas will accumulate inside the swimmer and drive the swimmer up (see Figure S4). In contrast, decreasing the applied electric field will allow gas to be released faster than it is produced, thus causing the swimmer to sink. The balance of both forces controls the buoyancy-driven motion (see Figure S5 and Video 1 in SI).

In order to not only control the motion in the vertical direction, but also along a horizontal axis, an alternative swimmer design has been adapted. It is based on a polymer cube (d) with two metal wires, one along the vertical axis and one along the horizontal axis. The latter wire is in addition modified at its both extremities with two tilted polymer plates (e) (Figures 3E, 3F). A platinum mesh is attached to the bottom of the vertical electrode to facilitate water reduction due to the electrocatalytic properties of platinum. Two orthogonal electric fields were applied using two sets of voltage generators. At the horizontal bipolar electrode, H_2 and O_2 are formed at the two extremities and since the volume of produced H_2 is twice that of O_2 , the resulting force will push the swimmer from the H_2 production side towards the O_2 production side³⁴. The tilted plates increase the surface of interaction with the bubbles in order to generate an optimized action/reaction force for the horizontal translation. A movement in both horizontal directions can be achieved by inverting the polarity of the electric field (see Videos 2 and 3 in SI).

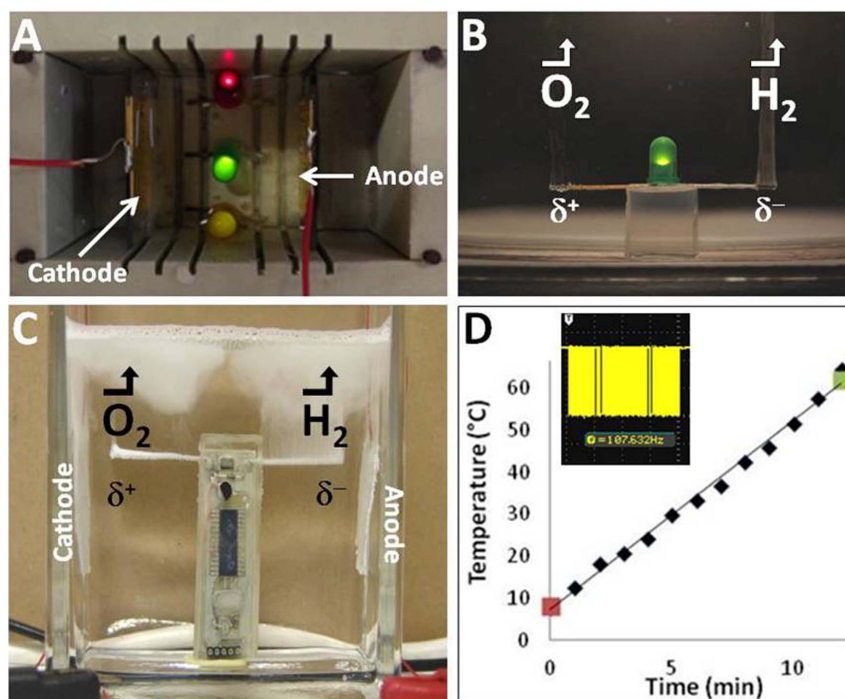


Figure 2 | Examples of contactless powered electronic devices (A) Contactless simultaneous powering of three LEDs immersed in an aqueous solution and exposed to the electric field between the feeder anode and cathode (top view of the cell) (B) Side view of one LED immersed in water and exposed to an electric field. The induced polarization (δ^- and δ^+) leads to H_2 and O_2 evolution at the electrical input and output of the diode. The resulting local current powers the LED which then emits light (C) Electronic circuit of a temperature sensor (for details see Figure S2) immersed in water and exposed to the electric field between cathode and anode. The polarization leads to the generation of gases and a local electric current which powers the circuit. An integrated emitter allows communication with a receiver placed outside of the water reservoir. The emitted infrared signal encodes the information about the local temperature. (D) Contactless measurement of the temperature in the solution as a function of time. Inset: Received signal from the temperature sensor. The number of pulses is proportional to the temperature in the solution (see Figure S3). Red and green squares represent the initial and final temperature respectively measured with a conventional thermometer.

The hydrodynamic behavior in the vertical direction of the rocket and the cube swimmers is different. For the rocket swimmer the characteristic rising and sinking periods are directly controlled by

the applied electric field (here from 0 V cm^{-1} to 2.9 V cm^{-1}) and the diameter of the gas release hole. Figure S5B shows a certain phase shift between the changes in applied potential and the resulting change in height, which corresponds to the time necessary for the accumulation or release of gas. The vertical speed of the rocket swimmer increases linearly with time, because the equilibrium between gas production and release leads to a steady state amount of gas accumulated inside the rocket and therefore to a constant acceleration. On the other hand, the cube tends to constantly accumulate increasing amounts of bubbles underneath the swimmer as a function of time, which corresponds to a gradual increase in driving force and thus an increasing acceleration (see Figure S5C).

As the evolution of hydrogen and oxygen at the two opposite sides of the swimmer during the bipolar electrochemical experiment is intimately related to a shuttling of electrons through the object, a local electric current exists that can be used for the simultaneous operation of an electronic device. As proof-of-principle we have chosen LEDs ((c) in Figures 3D and 3H) to confer an additional functionality to the object, thus leading to the first example of *e*-swimmers based on this concept.

Figures 3C and 3G show such light emitting swimmers. The vertical velocity of the light emitting rocket increases again linearly with time (Figure 4A). For the light emitting cube swimmer, two LEDs with different colors were integrated in opposite working directions in order to track the polarity of the applied horizontal field (Figure 4B). When the polarity of the horizontal electric field is inverted during the upward motion, not only the direction of motion changes, but also the active LED (and color) changes (Figure 4C and Video 5 in SI). It is clear that the motion in the horizontal direction is less spectacular and slower than in the vertical direction which is due

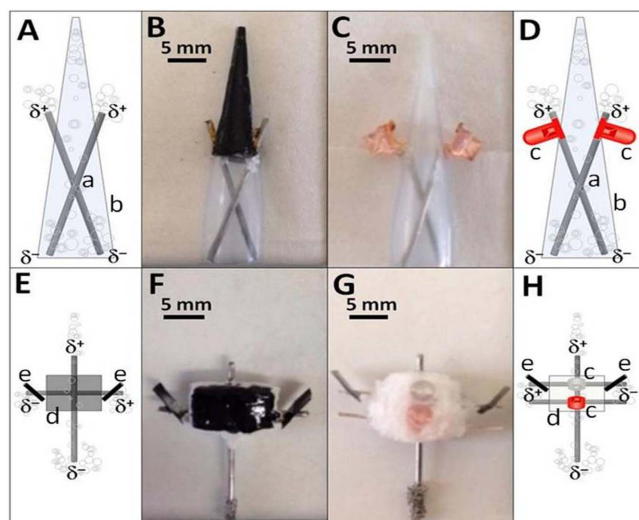


Figure 3 | Illustration of two different *e*-swimmer designs. (A) Scheme of the rocket swimmer; (B) Rocket swimmer; (C) Rocket swimmer with integrated LEDs; (D) Scheme of the rocket swimmer with integrated LEDs; (E) Scheme of the cube swimmer; (F) Cube swimmer; (G) Cube swimmer with integrated LEDs; (H) Scheme of the cube swimmer with integrated LEDs. a: metal wire; b: plastic cone; c: LED; d: polystyrene cube; e: polymer plates.

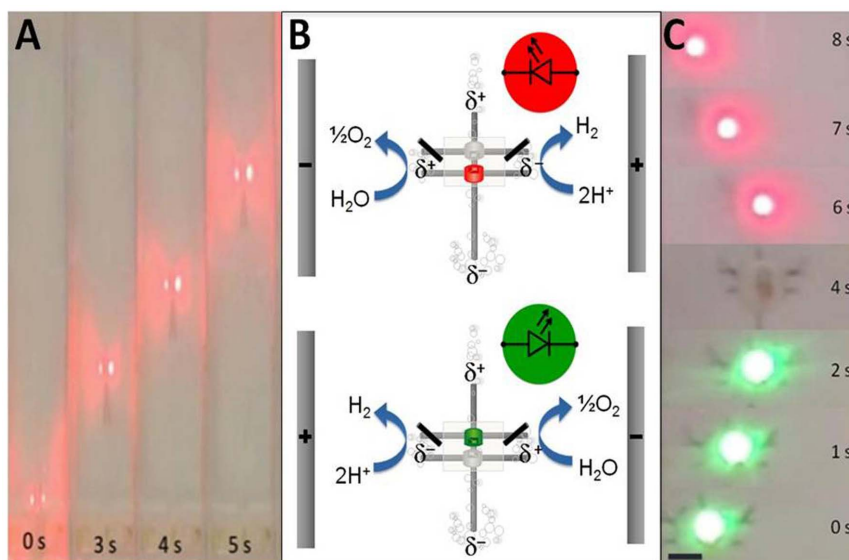


Figure 4 | Motion of light emitting *e*-swimmers (A) Series of optical frames showing the rocket LED-swimmer of Figure 3C during the rising in the electrochemical cell (see also Video 4 in SI). (B) Scheme of the color switching when inverting the direction of horizontal motion. Two LEDs with opposite orientation and different colors are incorporated into the swimmer. Inversion of the electric field leads to the switching on of one LED and the switching off of the other one. (C) Series of video frames showing the cube LED-swimmer during the vertical rising while changing the polarity of the horizontal electric field (11 V cm^{-1} in *z*-direction and 44 V cm^{-1} in *x*-direction); inversion of the polarity at 4 s leads to a change in color and of the direction of the horizontal motion (see also Video 5 in SI). Scale bar: 5 mm.

to the partial lack of buoyancy forces, but nevertheless it is possible to change the direction of motion by a change in polarity of the electric field.

Discussion

From the obtained results one can conclude that the intrinsic symmetry-breaking of bipolar electrochemistry can be exploited to simultaneously propel and power moving electronic components in an aqueous open space configuration. In a first proof-of-principle experiment LEDs have been integrated into the *e*-swimmers so that their motion can be tracked *via* light emission. It was also possible to operate much more sophisticated electronic devices by bipolar electrochemistry. Designing swimmers with different gas release kinetics allows precise control of the motion under otherwise identical conditions (see also Video 6 in SI). The most appealing aspect is the possibility to generate in a wireless way a localized electric current through the object that is strong and stable enough to allow the operation of electronic devices. However the conversion efficiency of such a set-up is rather low. For driving the electronic function one has to keep in mind first of all that in such a bipolar electrochemical set-up a large fraction of the current flows through the solution around the device and not through the device. In the present case this Faradaic efficiency can be estimated by dividing the current flowing through the swimmer (min. 1 mA in order to switch on the LED, max. 20 mA, otherwise the LED is destroyed) by the total current flowing through the cell in one direction (ca. 500 mA). With these values in mind one can calculate that the efficiency must be between 0.2% and 4.0%. The second important type of efficiency is the propulsion efficiency. This one can be calculated by the ratio of the electric energy that goes through the swimmer compared to the final kinetic energy of the object. The electric power which goes through the swimmer can be obtained by multiplying the current flowing through the bipolar object (min. 1 mA; max. 20 mA) with the voltage necessary to drive the LED (around 1 V) or the minimum voltage difference to obtain water splitting at the two ends of the swimmer (1.4 V). A value between 1 mW and 30 mW is obtained. Another way to estimate more precisely the current through the swimmer is to measure the volume of gas produced per time unit

at the swimmer electrodes. For this, an experiment has been performed that allows quantifying roughly the gas evolution triggered by the current passing through the swimmer (see Video 7 in SI). As can be seen from the video, the volume of hydrogen gas produced and trapped in a cap that is positioned over the left electrode of the swimmer is higher than the volume of oxygen gas (right side) due to the intrinsic stoichiometry of the water splitting reaction. A volume of approximately 0.13 cm^3 is obtained after a reaction time of 180 s. Taking into account the molar volume of the gas, $6 \cdot 10^{-6} \text{ mol}$ of H_2 are produced. Converting this value into a flow of charge leads to roughly 6 mA of current flowing through the object, which is in between the two current limits mentioned above. One can therefore assume that the power input into the swimmer is around 6 mW. Integrating this power over the duration of the experiment, leads to a total energy of 1 W.s (= 1 J) that has been injected. On the other hand the speed of the swimmer observed in the video is roughly 3 cm/s. This speed can be controlled by several parameters, such as:

- the amplitude of the electric field,
- the composition of the electrolyte,
- the design of the swimmer,
- the weight of the swimmer
- and also the presence of surfactant which changes the size of the produced bubbles.

With a typical swimmer mass of 0.5 g and the above mentioned speed, this leads to a kinetic energy of 0.2 μJ . The ratio of both values indicates that the conversion efficiency is around 0.00002%. If one combines this efficiency with the Faradaic efficiency as calculated above (around 1.2% assuming that 6 mA out of the 500 mA are going through the swimmer) then the total efficiency ends up to be $3 \cdot 10^{-9}$ or $3 \cdot 10^{-7}\%$. These values appear to be extremely low, but are in very good agreement with the values that Mallouk and coworkers³⁷ have reported for other bubble propelled micromotors (conversion efficiencies of the order of 10^{-9}).

On the other hand, the intrinsic advantage of the proposed concept is that many objects can be addressed simultaneously without physical contact to the feeder electrodes. Besides LEDs it is possible to power integrated circuits with much more complex features, enab-



ling the design of multi-functional *e*-swimmers. An extension of this work to 3D-controlled motion can be imagined by integrating an additional set of electrodes that enable *in fine* a total control of the upward, downward and sideward motion. This will allow for example the development of roving, wireless sensor chips, able to carry out several tasks and at the same time to ensure contactless interfacing with external electronic devices. However down-sizing the swimmers to very small scales, which means performing experiments at low Reynolds numbers, seems unrealistic due to the intrinsic limitations of bipolar electrochemistry related to equation 1. This means when the characteristic size *l* decreases, the electric field *E* has to increase proportionally in order to produce the same polarization voltage ΔV that allows electrolysis of water. Therefore, and also due to the characteristic size of microelectronic devices, the smallest dimensions of this type of swimmers should be in the sub-mm range in order to ensure reasonable experimental conditions.

We hope that the presented new concept will enrich the global spectrum of miniaturized robotic systems and allow further progress in this area, especially with respect to the wireless transmission of energy to and from moving objects, which recently is attracting increasing attention³⁸.

Methods

Chemicals. Solutions were prepared using Milli-Q water (resistivity = 18.2 M Ω .cm), HCl (Alfa Aesar, 36%) and hydroquinone (Aldrich-Chemie, analysis grade). When necessary, NaCl (Fluka analytical) was introduced so that the current flowing through the swimmer is not limited by the overall conductivity of the solution. SDS surfactant (Sigma, $\geq 98.5\%$) was added to the solutions in order to promote the formation of homogeneous bubbles and to facilitate their continuous release. The composition of the electrolyte has an impact on the efficiency of a bipolar electrochemical experiment in general and on the swimmer motion in particular. Higher electrolyte concentrations will slow down the motion. However it has been recently shown in a different context that bipolar electrochemistry is even possible in ionic liquids³⁹, which exhibit by definition a very good conductivity.

Experimental set-up. Electronic devices or *e*-swimmers were operated in a water containing reservoir, equipped with electrode arrays allowing to apply to the solution a horizontal and a vertical electric field, either separately or simultaneously (Figure S1). Two similar systems were built in order to generate motion, both of them were composed of two transparent plastic boxes, one inside the other. The smaller box was fixed inside the bigger reservoir in such a way that some space was left with respect to the bottom of the reservoir. Two couples of Pt electrode arrays were fixed around the smaller cell, one in *z*-direction, one in *x*-direction in order to generate an electric field as homogenous as possible, while maintaining Faradaic current as low as possible. The first set-up had dimensions for the outer cell of 17.2 \times 10.7 \times 2.4 cm, and for the inner cell of 12 \times 8.5 \times 1.2 cm. A second set-up had dimensions of 10 \times 25 \times 3 cm for the outer cell, and 7 \times 20 \times 1.5 cm for the inner cell. The cells were filled with a 10 mM aqueous SDS solution and the pH was set to 3.7 by adding a few drops of 1 M HCl. The values of the applied electric field were chosen as a function of the voltage drop necessary to drive either the electronic device, the swimmer or the combined *e*-swimmer. The motion was recorded using a digital camera.

The electric fields are generated by two types of equipment: one power supply able to generate a potential difference of up to 600 V and a maximum current of 1 A (Heinzinger PNC 600–1000), and a second one able to generate a potential difference of up to 350 V and a maximum current of 0.4 A (Fug MNC 140–350). The first one was used to generate the horizontal electric field, while the second one was used for the vertical electric field. The established fields are homogenous enough to allow a continuous motion of the swimmer. Typical current densities, calculated with respect to the geometric sections of the cell, are of the order of 0.05 A/cm² for both field directions.

The powered electronic parts are either commercial Light-Emitting Diodes (Radiospares) or a custom made electronic circuit comprising an integrated temperature sensor, an analog-digital converter and an infrared emitter (Figure S2). The whole electronic circuit is isolated from the aqueous environment by an epoxy sealing, only the two metal wires that are acting as bipolar electrodes are in contact with the solution and get polarized by the external electric field. The resulting electrochemical reactions lead to a local current flow through the electronic circuit, allowing its successful operation. The integrated temperature sensor generates an analog voltage proportional to the temperature of the solution. The microcontroller converts the analog voltage into a digital variable. The microcontroller controls also the infrared LED and emits every 500 ms pulses corresponding to the value of the digital conversion.

The swimmer design is based on the competition between the kinetics of gas generation and release. Changing this ratio in the case of the rocket type swimmer by increasing the opening at the top allows adjusting this equilibrium as illustrated in Figure S4.

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Author contributions

J.R., S.C., G.L., J.L., L.B., P.F. and A.K. discussed and designed the experimental set-up; J.S. developed the electronic device; J.R., S.C., J.L. and G.L. carried out the experiments; all authors contributed to the production of the manuscript.

Additional information

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