Supplementary Information

Soft Electrochemical Bubble Actuator with Liquid Metal Electrode Using an Embodied Hydrogel Pneumatic Source

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Figure S1. Relative bubble volume as a function of time. A voltage of 10 V is applied for 30 s to initially inflate the chamber and then the voltage was switched off. V(t) is the volume at time 't' and V_0 is the volume immediately after the voltage source is switched off. Pictures were taken every 30 mins and analyzed using Image J to get the dimensions of the bubble to evaluate volume.



Figure S2. Role of the distance between the electrodes. (A) The current generated and (B) the bubble volume as a function of the distance between the electrodes. A voltage of 5 V is applied in each case for 30 s and the current going through the system and the bubble volume were recorded at 30 s.

Role of the distance between the electrodes: We varied the distance between the electrodes from 2.5 mm to 10 mm to elucidate the role of device architecture in output bubble volume. Increasing the distance increases the overall resistance of the device through the gel between the electrodes. As expected, we observe a decrease in the current and the bubble volume with increasing distance between the electrodes as shown in Figures S2A and S2B respectively.



Figure S3. (A) Bubble volume as a function of the voltage applied. The bubble volume was recorded at 30 s in each case while a corresponding voltage was applied. The red line denotes the fit, the equation has an x-intercept of 3 V. When a voltage of (B) 1 V, (C) 2 V, and (D) 3 V was applied for 30 s, the bubble does not inflate significantly, this explains the intercept.



Figure S4. Bubble actuator with different counter electrodes. (A) A carbon rod of 1mm diameter. The carbon rod was removed while taking the picture. However, removing the carbon rod left a trace in the gel and the red dashed arrow points to the trace where the carbon rod was inserted. (B) Carbon felt. Both samples are 3 cm long initially.

The device shown in Figure S4A consists of three LM droplets connected by a thin copper wire and the counter electrode (1 mm diameter carbon rod) is placed on the right side of the sample. The liquid metal droplet closer to the counter electrode generated larger and faster actuation than those that are further apart (Figure S2). Whereas in the device shown in Figure S4B, a carbon felt is used as a counter electrode that is stuck on the whole bottom side of the gel, which is equidistant from the two LM droplet electrodes. As the counter electrode is equidistant from both droplets, both bubbles expanded to nearly the same size at the same time scales.

Figure S4 indicates that the position of the electrodes could dictate the curvature generated in the gel. For example, in Figure S4A, the LM droplet closer to the counter electrode forms the largest bubble whereas, in Figure S4B, uniform and symmetrical bubbles inflate to the same volume. Hence, this system presents a wider scope for controlled actuation.



Figure S5. Bubble volume as a function of time at various input voltages (5 V, 10 V, and 15 V).



Figure S6. (A) Bending angle of the actuator shown in Figure 3B of the manuscript as a function of time and the method used to estimate the bending angle (B).



Figure S7. Side view and top view of the bubble. Bubble volume is estimated using the volume equation for an ellipsoid, where *a* was the x-radius of the ellipsoid and *b* was the y-radius of the ellipsoid, and *c* was the height of the ellipsoid.

Empirical data predicting equations

(i) Pressure approximation

The chamber expands when the pressure generated by the gas formed inside the chamber wall overcomes the elastic work required to expand the hydrogel ($E(2\pi rt)$), and the outside atmospheric pressure exerted on the chamber walls ($P_{out}(\pi r^2)$). Here, r is the radius of the bubble, t is the thickness of the hydrogel bubble membrane, E is the modulus of the gel, and P_{out}

is the pressure outside the bubble. Substituting the values for E = 40 kPa (measured experimentally via stress-strain measurements in tension on an Instron extensometer), $P_{out} = P_{atm} = 101$ kPa, r = 1 cm (approximately the maximum volume observed in our experiments), t = 0.15 cm in the expressions above suggests that the pressure exerted by the atmosphere is about two orders of magnitude higher than the elastic work required. Thus, we assume pressure inside the bubble does not differ significantly from the outside atmospheric pressure.

(ii) Current and volume prediction equations

Current decay for a constant step voltage applied using Randles equivalent circuit is

$$i(t) = A + Bexp(-t/\tau)$$

where $A = V_{app}/(R_s + R_p)$, $B = V_{app}R_p/R_s(R_s + R_p)$, τ is the time constant ($R_sR_pC_d/(R_s + R_p)$), V_{app} is the voltage applied to the system, R_s is the solution resistance, R_p is the polarization resistance, and C_d is the electrical double layer capacitance.

By fitting the data in Figure 2 using the equation above, we get an R² of 0.986. Values of A, B, τ and the corresponding R_s , R_p , C_d are provided in the table below. We use the Simple Fit tool in Origin software to fit the data.

Fitting Parameter	Value	Circuit Element	Value
А	0.019	$R_s\left(\Omega\right)$	78.135
В	0.045	$R_p\left(\Omega ight)$	185.023
τ	35.773	C_d (F)	0.65

Table S1. Fitting parameters for Equation 4.

$$Volume(t) = D \times \int_{0}^{t} i(t)dt$$

We estimate volume using,

where D is equal to $RT/nFP_{atm.}$

$$i(t) = 0.019 + 0.045 \times exp(-t/35.773)$$

$$\int_{0}^{t} i(t)dt = 0.019 * t - 1.60979 * exp(-0.027954 * t) + 1.60979$$

Volume
$$(t) = D * \int_{0}^{t} i(t)dt$$

Volume (t)
=
$$155.73686 \times (0.019 \times t - 1.60979 * exp(-0.027954 \times t) + 1.60979)$$

 mm^3

(ii) Circuit element estimates reported in Table S1

We conducted impedance spectroscopy on the device (Figure S7A) and fit the data using a Randles equivalent circuit (REC) Figure S7B), to evaluate the circuit elements R_s , R_p , and C_d . We found them to be 27 Ω , 2340 Ω , and 2.8 μ F respectively (Figure S7). The impedance was conducted at 0 V with respect to the open circuit voltage and by applying a 10 mV AC voltage. Although the R_s generated by the fitting parameters (Table S1) is on the same order of magnitude, other elements (R_p , C_d) generated by the fitting of the current decay (Table S1) differ significantly from those evaluated from the impedance data. Thus, rather than considering Equation 4 as a Randle's circuit, we simply consider it an exponential decay.



Figure S7. Evaluation of circuit elements. (A) Impedance spectroscopy of the LM-hydrogel device. Blue data points indicate the experimental results whereas the red data points indicate the fit generated using Randles equivalent circuit (REC) (B). (C) Circuit parameters obtained from the impedance data fitting using REC.

The discrepancies in the fitting parameters while using a Randle's circuit model could arise from several things in our system such as the (i) increase in the cell resistance with time due to the oxide formation at the anode, (ii) decrease in the contact area between the electrode and the gel due to the high surface tension LM forming a sphere upon reduction at the cathode, (iii) depletion of reactants in the vicinity of the electrodes and diffusion constraints, (iv) the REC circuit does not capture all the impedance data; thus, the values of circuit elements might not be accurate.

Despite all the deviations that arise from simplifying this complex system to a straightforward electrochemical system (Randles equivalent circuit), we use this simple model because it allows us to capture the exponential decay in current and serves the purpose of providing mathematical relationships that describe voltage and current as a function of time. We include the Randles circuit analysis here to show that we attempted to use a theoretical framework to predict current versus time in case it aids future researchers.

To determine D, we substituted $R = 82057 \text{ mm}^3$ atm K⁻¹ mol⁻¹, T = 295 K, n = 2, F = 96485 C mol⁻¹, $P_{atm} = 1.0$ atm results in $D = \frac{RT/nFP_{atm}}{126}$. However, the empirical value from the best fit of the data shown in Figure 2C using D as a fitting parameter result in D = 156. This small discrepancy could be due to (i) Challenges associated with estimating the bubble volume. We assume the bubble is an ellipsoid. We measure the bubble volume by measuring the diameters of the ellipsoid manually from the side and top view pictures using the Image J software. Thus, there could be room for overestimation. (ii) The initial LM pocket could play a role in resulting in higher experimental values than the theoretical prediction. The initial volume of the cavity is defined by the metal and therefore the initial volume requires no gas. In addition, the initial cavity is a rectangular prism, rather than an ellipsoid.

Supporting Videos

S1 – The gels are 5.5 cm long, the input voltage is 15 V, and the real time length of the movie is 20 s. The SI video is played at $4 \times$ speed.

S2 – The gel is 3 cm long, and the input voltage is 5 V. The SI video is in real time.

S3 – The carbon felt seen in the video is 5 cm long. The input voltage is 10 V, and the video is in real time.

S4 – The input voltage is 10 V, and the SI video is played at 8× speed. The voltage is on for 30 s before lifting the ping pong ball (3.8 cm wide).

S5 – The input voltage is 10V and the SI video is played at 4x. The voltage is on for 16 s before lifting the 20 ml glass vial (2.73 cm wide).