"Fluidic Batteries" as Low-Cost Sources of Power in Paper-Based Microfluidic Devices

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Materials

Sodium nitrate, aluminum metal, silver metal, β -D-galactosidase, silver nitrate, dimethyl sulfoxide, anhydrous aluminum chloride, and mono- and di- basic sodium phosphate were purchased commercially and used without further purification. The paper used was Whatman Chromatrography Paper Grade 1, and the tape was Ace Hardware Plastic carpet tape (part # 50106). All solutions were prepared using distilled water. The technicloth used was TechniCloth[®]11 (TX[®]1109, Wipers (Blend), 45% polyester/55% cellulose, 23 cm × 23 cm). The copper tape (6.3 mm wide, 0.04 mm thick copper, product # 16072) and XYZ conductive tape (12.7 mm wide, 0.127 mm thick, product # 16081-82) were purchased from TedPella.com. The red LED (item # SML-211UT) was purchased from Farnell, and the UV LED (Model # NSSU123T) was purchased from Nichia Corporation.

General Procedure for Fabricating the Devices:

Patterning paper

The paper was patterned according to the procedure described by Noh.¹ The only variation made was heating the paper for only 105 seconds rather than 120 s after printing wax on the paper using a wax printer.

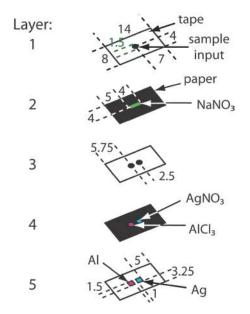
Patterning tape

The tape was patterned according to the procedure described by Noh.¹

Fabricating 3D µPADs

The general procedure for assembling the devices is described in Refs. 1 and 2. Specific variations of that procedure are described below. For example, the salt bridges were prepared by spotting 0.375 μ L of saturated aqueous NaNO₃ solution twice into a designated hydrophilic region of paper, and by drying the wet paper under vacuum for 30 min after each application. The electrolytes, 0.25 μ L each of 1 M AlCl₃ and 3 M AgNO₃, were spotted and allowed to dry under vacuum for 30 min. After each layer was dried, the layers were cut to size using scissors. The holes in the inner layers of tape in the devices were filled with technicloth that had dimensions equal to the size of the holes unless otherwise noted. The assembled 3D μ PADs were compressed using a 4.9 kg weight for approximately 15 minutes. The electrode layer contained pieces of aluminum and silver metal (0.25 mm thick) cut to dimensions equal to the size of the holes in the tape. After placing the metal into the holes, the holes were covered using a piece of copper tape.

Single Cell Battery

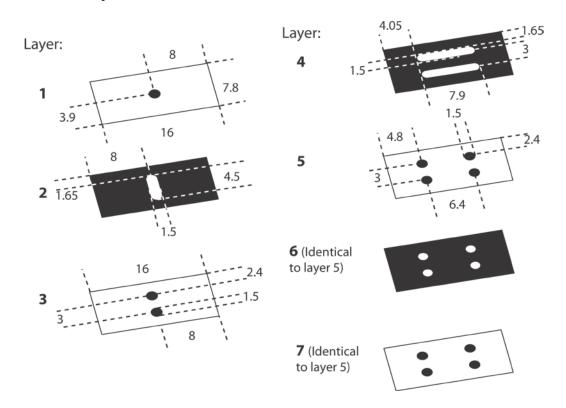




Fabrication of a single cell battery.

This device was designed as shown in Figure S1, with the salt bridge in layer 2 and the electrolytes in layer 4, which were prepared as described above. Aluminum and silver metal pieces were placed in the holes in the layer 5. Copper tape was placed over the pieces of metal to hold them in place. When

testing this design, a multimeter was connected (using alligator clips) to the pieces of copper tape, and 5 μ L of distilled water was deposited onto the sample input region in layer 1.

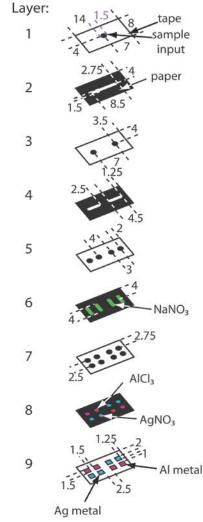


Two-Cell Battery

Figure S2. Expanded view of a 2-cell battery. All values shown are in millimeters. The device is 16-mm wide × 7.8-mm long. A color-coded version of this device is shown in Figure 1.

Fabrication of a 2-cell battery.

The device was designed as shown in Figure S2, with the salt bridges in layer 4 and the electrolytes in layer 6, which were prepared as described above. Aluminum and silver metal were placed in the holes in layer 7. Copper tape was placed over the pieces of metal, acting as an electrode and a connection between the two cells, as shown in Figure 1. This design was tested by connecting the copper tape to a multimeter in the method shown in Figure 1a or 1b, and by depositing 5 μ L of distilled water to the sample input region in layer 1.



Four-Cell Battery

Figure S3. Expanded view of a 4-cell battery. All values shown are in millimeters. The device is 14-mm wide × 8-mm long.

Fabrication of a 4-cell battery.

The device was designed as shown in Figure S3, with the salt bridges in layer 6 and the electrolytes in layer 8, which were prepared as described above. Aluminum and silver metal were placed in the holes in layer 9. Copper tape was placed over the pieces of metal, acting as an electrode and a connection between the four cells. This design was tested by connecting the copper tape to a multimeter and by adding 10 μ L of distilled water to the sample input region in layer 1.

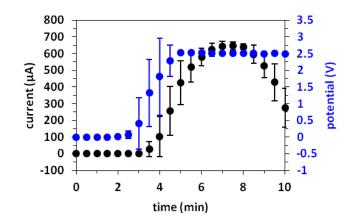


Figure S4. Power output for the four-cell battery after adding 10 μ L of distilled water at 25 °C. The potential is the average of four trials and the current is the average of 5 trials. The error bars are the standard deviations from these averages.

Table S1. Current and potential data for the four cell battery as shown in Figure S4. (Note: measurements of potential were repeated four times, while measurements of current were repeated five times.)

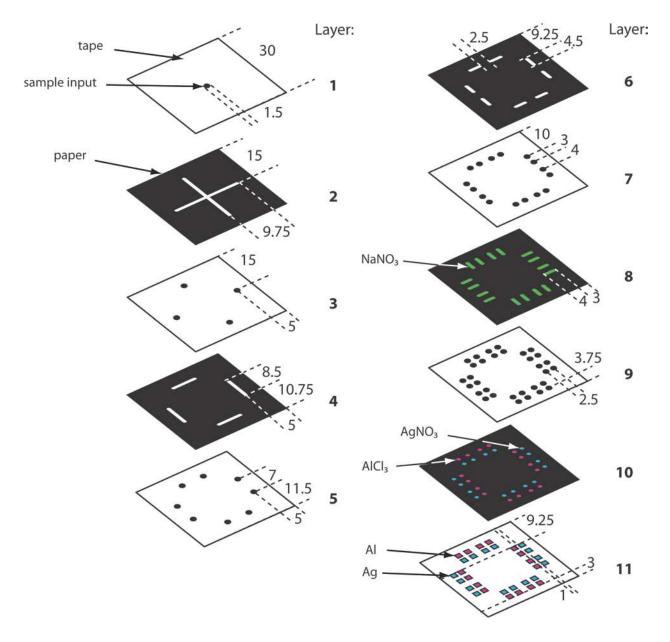
Time (min)	Current (µA)	Current Standard Deviation	Potential (V)	Potential Standard Deviation
0	0.0	0	0.000	0.000
0.5	0.0	0	0.000	0.000
1	0.0	0	0.000	0.000
1.5	0.0	0	0.000	0.000
2	0.0	0	0.018	0.029
2.5	0.0	0	0.078	0.113
3	0.8	1.8	0.408	0.763
3.5	26.4	45.6	1.318	1.010
4	103.8	120.2	1.805	1.161
4.5	256.8	147.5	2.285	0.464
5	425.0	132.6	2.525	0.047
5.5	519.0	91.4	2.525	0.038
6	577.8	52.6	2.520	0.029
6.5	623.4	39.9	2.518	0.017
7	641.6	30.7	2.503	0.017
7.5	645.8	24.4	2.515	0.017
8	641.0	17.1	2.808	0.021
8.5	596.0	56.2	2.503	0.026
9	528.2	72.9	2.495	0.024
9.5	427.2	111.2	2.500	0.018
10	276.0	116.2	2.498	0.021

Measuring the relative intensity of light produced by the red LED in the four cell design shown in Figure 1d.

The light output of the LED was measured as resistance using a photoresistor. The LED was attached to the fluidic battery, and the photoresistor was placed approximately 2.5 cm above the LED. The resistance was measured using a multimeter. Both the battery and the photoresistor were adhered to a piece of glass, and a small box was placed over them to remove most of the ambient light. A piece of black canvas (folded so that it was 4 layers thick) was placed over the box to remove the rest of the ambient light. During testing, 10 μ L of distilled water was added to the inlet of the device outside the box, and then the device was placed within the box immediately.

Table S2. Resistance measurements obtained using a photoresistor placed over the LED in the four-cell battery. All values are in K Ω . The error is the standard deviation of the five trials. The intensity of light emitted by the LED is approximately inversely proportional to the resistance of the photoresistor.

P=======							
Time (min)	Trial 1	Trial 2	Trial 3	Trial 4	Trial 5	Average	Error
0	2000	2000	2000	2000	2000	2000	0
0.5	2000	2000	2000	2000	2000	2000	0
1	2000	2000	2000	2000	2000	2000	0
1.5	2000	2000	2000	2000	2000	2000	0
2	2000	2000	2000	2000	2000	2000	0
2.5	2000	2000	2000	2000	2000	2000	0
3	2000	2000	2000	2000	2000	2000	0
3.5	2000	2000	391	469	2000	1372	860
4	2000	2000	220	292	2000	1302	956
4.5	280	504	130	152	890	391	316
5	133	135	104	110	230	142	51
5.5	89	82	90	96	119	95	14
6	74	61	83	170	95	97	43
6.5	68	53	77	250	83	106	81
7	69	48	78	235	65	99	77
7.5	56	46	79	198	57	87	63
8	89	44	77	202	53	93	64
8.5	56	43	82	180	53	83	56
9	54	44	84	180	54	83	56
9.5	82	43	81	270	66	108	92
10	58	43	87	176	60	85	53
10.5	53	44	89	181	55	84	57
11	56	45	93	155	54	81	45
11.5	56	46	95	143	55	79	40
12	58	48	101	220	59	97	72
12.5	60	50	103	328	58	120	118
13	60	51	109	350	60	126	127
13.5	62	54	122	299	58	119	104
14	63	56	153	250	65	117	84
14.5	64	59	285	262	62	146	116
15	66	63	370	181	63	149	134



16-Cell Battery

Figure S5. Expanded view of a 16-cell battery. Every layer of the device is completely symmetric and all values shown are in millimeters. The device is 30-mm wide × 30-mm long.

Fabrication of a 16-cell battery.

The device was designed as shown in Figure S5, with the salt bridges in layer 8 and the electrolytes in layer 10, which were prepared as described above. The holes in layer 11 were filled with pieces of Al and Ag as shown in Figure S5. These metal pieces were then covered with pieces of copper tape, which also connected the cells into four sets of four cells in parallel. Each of these sets of cells in parallel were connected in series using the copper tape.

24-Cell Battery

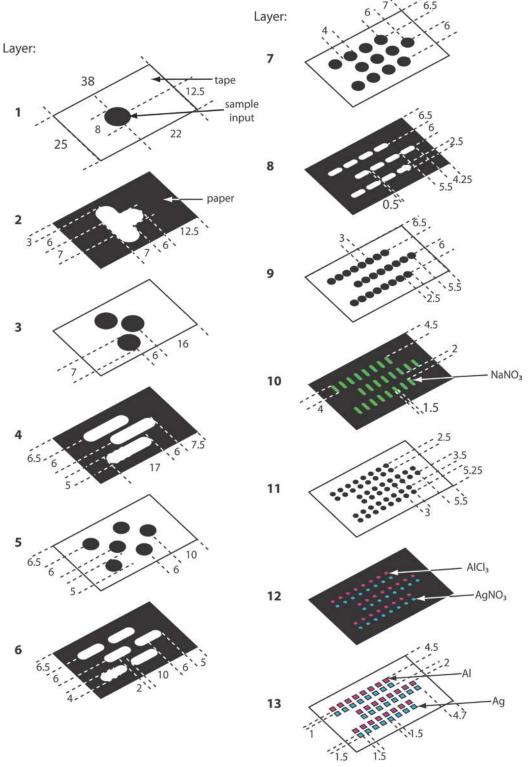
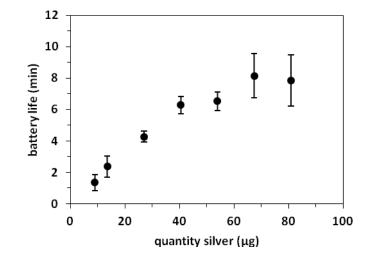


Figure S6. Expanded view of a 24-cell battery that includes wide fluidic channels. All values shown are in millimeters. The device is 38-mm wide × 25-mm long.

Table S3. I_{sc} and V_{oc} for the device shown in Figure S6. This design was tested by adding 500 μ L of distilled water to the sample input region and by monitoring the output using a multimeter. Measurements of current were repeated five times on five different devices, while measurements of potential were repeated three times on three different devices. Current and potential could not be measured simultaneously and continuously on the same device.

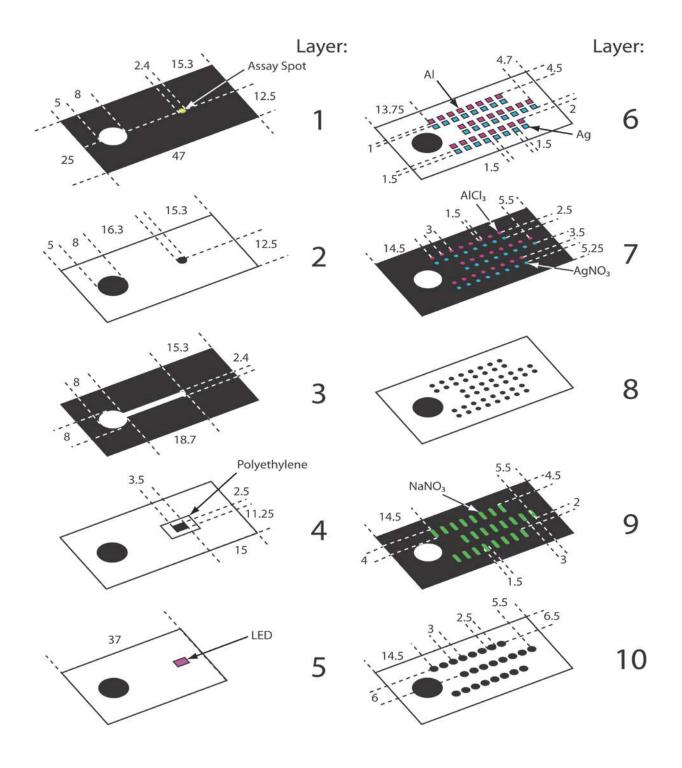
Time (min)	Current	Current	Current	Current	Current	Potential	Potential	Potential
	(mA)	(mA)	(mA)	(mA)	(mA)	(V)	(V)	(V)
	Trial 1	Trial 2	Trial 3	Trial 4	Trial 5	Trial 1	Trial 2	Trial 3
0	0	0	0	0	0	0	0	0
1	0.92	1.38	0.70	2.86	3.43	3.71	3.70	3.76
2	2.23	1.58	1.70	3.90	4.43	3.74	3.78	3.76
3	2.2	1.86	1.88	3.20	3.08	3.70	3.75	3.76
4	2.05	1.77	1.68	1.46	0.74	3.69	3.75	3.77
5	1.56	1.32	1.72	0.70	0.69	3.64	3.74	3.77
6	1.18	1.27	1.60	0.30	0.21	3.67	3.75	3.74
7	0.70	1.20	1.18	0.23	0.20	3.64	3.72	3.78
8	0.82	0.99	0.84	0.22	0.19	3.60	3.68	3.75
9	0.71	0.69	0.75	0.22	0.13	3.54	3.74	3.73
10	0.77	0.54	0.68	0.21	0.64	3.49	3.72	3.75
11	0.75	0.66	0.45	0.21	0.73	3.47	3.61	3.71
12	0.47	0.42	0.30	0.16	0.31	3.55	3.55	3.66
13	0.25	0.38	0.24	0.13	0.21	3.50	3.53	3.63
14	0.25	0.30	0.14	0.13	0.18	3.40	3.58	3.63
15	0.24	0.26	0.14	0.13	0.18	3.32	3.60	3.56
16	0.20	0.27	0.13	0.11	0.17	3.15	3.51	3.49
17	0.18	0.27	0.13	0.11	0.16	3.12	3.47	3.47
18	0.17	0.26	0.13	0.11	0.11	3.18	3.48	3.42
19	0.17	0.24	0.13	0.09	0.14	3.09	3.40	3.42
20	0.16	0.18	0.13	0.10	0.12	3.10	3.31	3.38



Effect of the Quantity of Electrolyte on the Lifetime of the Battery

Figure S7. The effect of changing the quantity of electrolytes (on a per cell basis) on the battery lifetime. The device shown in Figure S3 was used for these experiments. We presume that the plateau effect at high concentrations of silver is caused by the solubility limit of silver salts in the small volume of water (~0.2 μ L) that reaches the electrolyte.

Fluorescence Assays



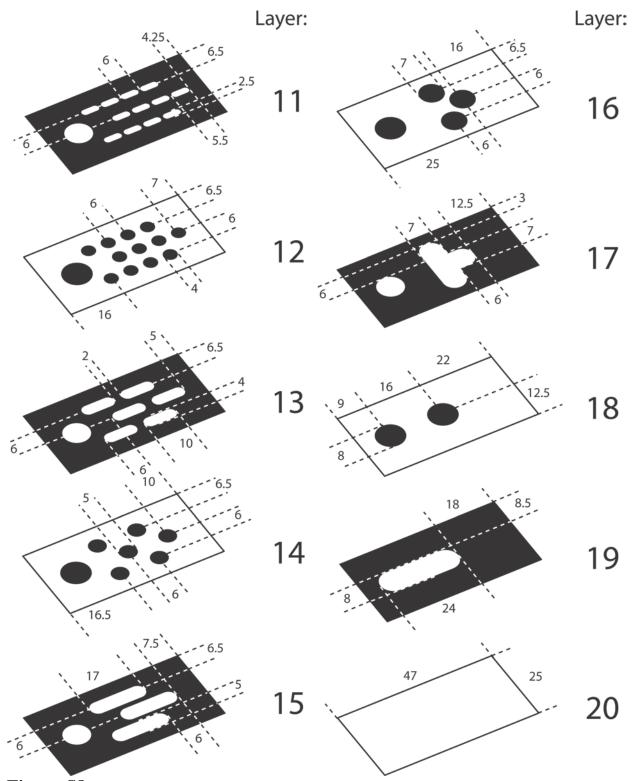


Figure S8. Expanded layers of the fluorescence assay device shown in Figure 2a. All values are in millimeters. The device is 47-mm wide × 25-mm long. A color-coded version is shown in Figure 2.

Fabrication of the fluorescence assay device.

The device was designed as shown in Figures 2a and S8, with the salt bridges in layer 9 and the electrolytes in layer 7, which were prepared as described above. The squares in layer 6 were filled with aluminum and silver metal. Copper tape was placed over the electrodes, connecting each cell down the row into three sets of 8 cells in parallel, and connecting each of the rows into series. Layer 5 and the UV LED were then added to the pre-assembled stack consisting of layers 6–20. The LED was connected to the electrodes using copper tape. Four replicates of layer 4 were then added to fully encase the height of the LED; a piece of polyethylene was placed over the LED hole to block liquid from contacting the LED.

The assay region in layer 1 was pre-spotted with 0.2 μ L of 78.6 mM compound 1⁴ in DMSO and was dried under vacuum for 1 h. To the same spot was added 0.2 μ L of 100 mM sodium phosphate buffer (pH = 7.2), and the layer was dried for another 30 min under vacuum. Layers 1–3 were then added to the stack, with the smaller hole in layer 2 filled by chromatography paper rather than technicloth.

Fluorescence assay for β -D-galactosidase.

Fluorescence assays were tested using β -D-galactosidase in distilled water. The entire bottle (4.37 mg) of the enzyme was dissolved in 1.9 mL of distilled water to provide a 5 μ M solution. The assay involved adding 350 μ L of the enzyme solution onto the input region on layer 1 of the device in Figure 2, and by allowing the sample to wick into the device. After 19 min, a picture was taken of the assay region using the camera installed on an iPhone4S. The intensity of the colored fluorescent output in the pictures was analyzed using Adobe®Photoshop®. In this proof-of-concept demonstration, the assay was run using both the 5 μ M solution of β -D-galactosidase and distilled water (i.e., 0 μ M β -D-galactosidase). The red channel in the histogram function in Adobe®Photoshop® was used to analyze the intensity of color in the photographs of the assays. The median intensity of the red value provided the following signals for the two assays: 147 for 0 μ M β -D-galactosidase and 47 for 5 μ M β -D-galactosidase.

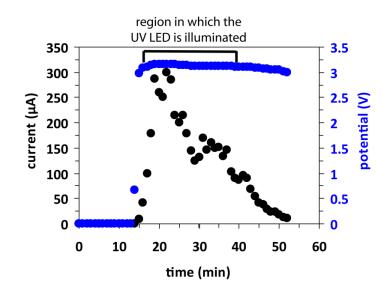


Figure S9. Power output for the fluorescence device in Figure 2a when the UV LED is connected to the fluidic battery. The experiment to measure the current and potential was stopped before the potential declined significantly.

References

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