Paper-based stacked reverse electrodialysis cells for energy generation from salinity gradient

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Introduction

The recently emerging microfluidic paper-based electrochemical devices (µPED) take advantage of both the capillarity of the paper and electrical detection in order to achieve low-cost, point-of-care and environmental-friendly analysis. The former eliminates active external pump requirement, while the latter allows for quantitative analysis. However, the electrical components still need external power sources to work, losing the portability advantage of paper-based analytical devices. A solution is to integrate the power source directly in the µPED to create a self-powered sensor [1].

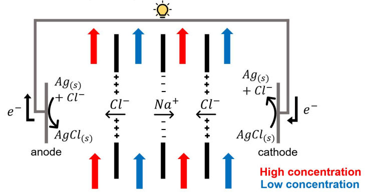
In this work, a paper-based stacked reverse electrodialysis (RED) cells is investigated as a potential power source. First, an innovative, accessible, and cost-efficient fabrication process is proposed to set up a paper-based stacked RED cell. Secondly, a microfluidic analytical model is investigated to characterize the cell efficiency. Finally, we successfully evaluate the power generation of the innovative paper-based RED device.

Operating Principle

Reverse Electrodialysis

RED principle convert **salinity gradient power** from electrolyte solutions of different concentrations to electrical energy through **ion exchange membranes** (IEM) and **redox reaction** [2].

Schematic principle :



Diffusion Potential :

$$E_{diff} = (2t_+ - 1)\frac{RT}{zF} ln \frac{\gamma_H c_H}{\gamma_L c_L}$$

High Concentration Compartment

Low Concentration Compartment

IEM dependence

Pros :
- Clean and sustainable energy (Blue Energy)
- Membrane-based technology
- No harmful byproduct

Cons :
- **Require active external pumping**
  → Energy cost and not portable

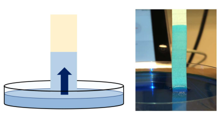
Paper-based RED

Paper-based RED cells take advantage of **spontaneous capillary flow** (SCF) to suppress the requirement of external pumping [3].

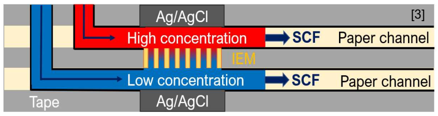
Pros :
- **Does not require external pump**
- Easy and low-cost fabrication
- Sustainable material (paper)
- Portable and Integrable to µPED

Cons :
- Difficult to model paper-based microfluidic
- Limitation in the flow rate
- Non-reusable device

Spontaneous capillary flow



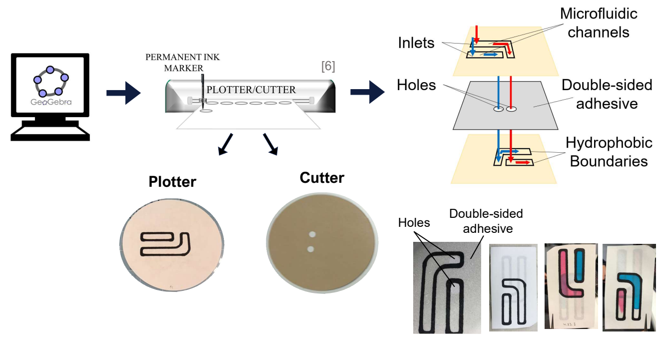
Stacked paper layers for RED



Fabrication

Microfluidic channels defined by applying a **permanent marker** with an automated plotter
→ hydrophobic boundaries confine and guide the liquid though the paper

3D structures achieved by **stacking** patterned paper layers
→ Handmade assembling by binding each layers with a **water-impermeable double-sided adhesive** patterned with an automated cutter



Pros :
- Good precision and homogeneity
- Good reproducibility
- Accessible and low-cost process
- Complex structure achievable

Cons :
- Time consuming (many steps)
- Difficult to design channels (curved edges)
- Difficult to accurately align the paper layers during the assembling

Microfluidic Design and Model

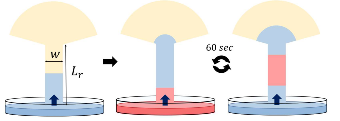
GOAL: Stationary flow rate of solutions close to the membranes = **Constant generation of power**

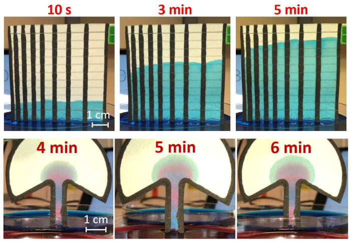
Quasi-Stationary flow rate (QSF)

Circular opening at the end of the channel

= Continuous increase of unwetted porous volume
→ compensates the continuous increase of viscous forces along the channel

→ **QSF**





Hydrophobic boundaries impact

Hydrophobic boundaries → decrease flow velocity (depending on width)

Microfluidic Model

QSF Model derived by combining :
- Mendez *et al.* model for fan-shaped channel [4]
- Hong and Kim model for channels patterned with hydrophobic boundaries [5]

Flow rate : $q_r = w\, b\, V_r$

↓

Flow velocity $V_r = \frac{\kappa_i P_{c,eff}}{\mu L_r}$

Paper

Length

↓

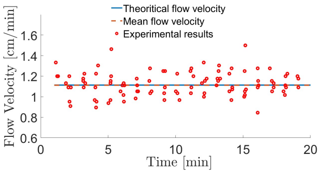
Effective capillary pressure $P_{c,eff} = P_c + \frac{K_H}{w}$

Paper

Hydrophobic material

Width

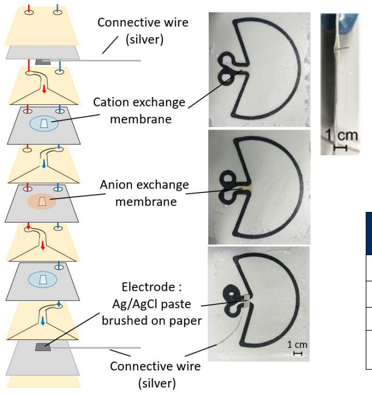
→ **QSF controllable with channel geometry**



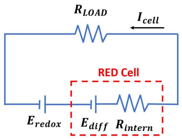
Limitations :
- Calibration of paper properties
- Strong assumptions (e.g. rigid porous media, saturated wetting front, …)

Paper-based stacked RED cell

Split view of stacked paper-based RED cell



Equivalent electrical circuit



Maximal power density :

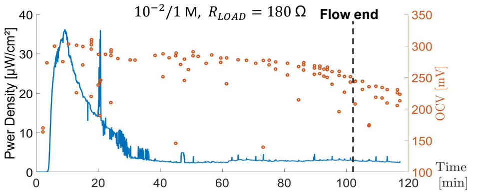
$$P_{max} = \frac{OCV^2}{4AR_{intern}}$$

OCV = Open circuit voltage
$= E_{diff} + E_{redox}$

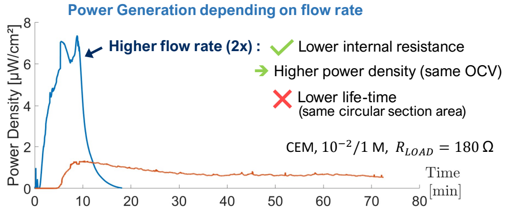
Properties of RED cell depending on IEM and concentration ratio :

RED Cell	Concentrations $[M]$	R_{intern} $[\Omega]$	OCV $[mV]$	$OCV_{100\%}$ $[mV]$	P_{max} $[\mu W/cm^2]$
CEM	$10^{-2}/10^{-1}$	331	65	109	3.19
CEM	$10^{-2}/1$	202	153	221	29
AEM	$10^{-2}/1$	364	2	0	0.002
Stacked (3 IEMs)	$10^{-2}/1$	178	262	442	95,33

Power generation of paper-based stacked RED cell



Power Generation depending on flow rate



Higher flow rate (2x) :
- ✓ Lower internal resistance
- → Higher power density (same OCV)
- ✗ Lower life-time (same circular section area)

CEM, $10^{-2}/1$ M, $R_{LOAD} = 180\ \Omega$

Conclusion

- **Accessible and low-cost fabrication process** successfully developed by combining an automated cutter/plotter and double-sided adhesive.
- **Quasi-stationary liquid flows** in the vicinity of the membrane successfully achieved and **modeled**.
- **Stacked paper-based RED cell** were successfully created and increase power density compare to previous work [3]. Last constraint : requirement of an additional component to compress the system. → Possible solutions: use of a laminator or creation of an external compressor device with 3D printing.
- **Power generation depends on liquid flow rate**, which can be **controlled** with channel geometry. Further research could investigate the optimization of the device.
- Life-time depends on flow rate and circular section area → power generation during times larger than one hour.

References

[1] A. M. López-Marzo *et al., Lab Chip* 16 (2010) 3150-3176.
[2] R. Ashu Tufa *et al., Applied Energy* 225 (2018) 290-331.
[3] H. K. Chang *et al., Lab Chip* 16 (2016) 700-708.
[4] S. Mendez *et al., Langmuir* 26(2) (2010) 1380-1385.
[5] S. Hong and W. Kim, *Microfluid Nanofluid* 19 (2015) 845-853.
[6] Nicolò Dossi *et al., Anal. Chem.* 89 (2017) 10454-10460.