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# Microbial Fuel Cell Technologies for Renewable Power and Biofuels Production From Waste Biomass

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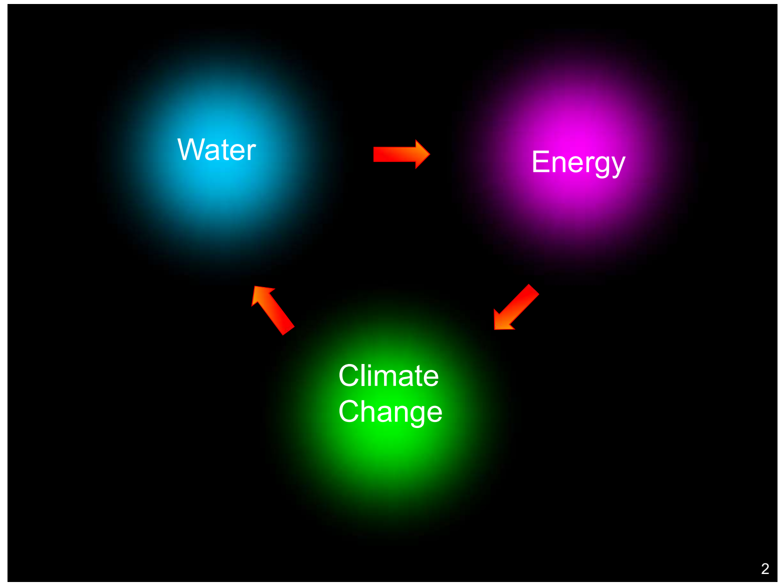
Engineering Energy & Environmental Institute

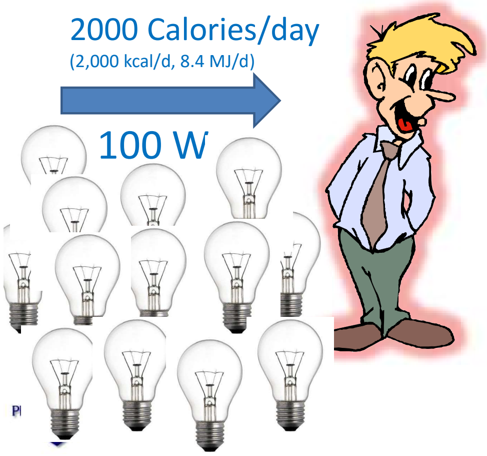





## Power Consumption by People

2000 Calories/day
(2,000 kcal/d, 8.4 MJ/d)

100 W





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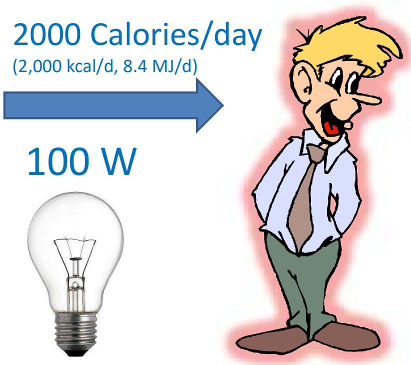

25 W "down the drain" (2 MJ/d)

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## Energy & The Water Infrastructure

- **Annual energy used for the water infrastructure**
  - 30 GW (USA), or 5-6% of all electricity generated
- **Energy USED for wastewater treatment**
  - 15 GW (USA)
  - **0.6 kWh/m³** (range: 0.12 to 1-2 kWh/m³ )
- **New energy SOURCE? (waste)water**
  - Domestic & Industrial wastewaters contain **17 GW** (USA)
  - Domestic wastewater contains (in the organic matter) about **2-5 kWh/m³**; or 4 - 10 times that needed using conventional treatment!

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## New Energy Sources Available using Microbial Electrochemical Technologies (METs)

- **Wastewater** : Organic matter in water (USA)
  - **17 GW** in wastewater
  (Save 45 GW energy/yr used + produce 17 GW = 62 GW net change)
- **Cellulose Biomass Energy**: Get biomass→ water
  - **600 GW** available (based on 1.34 billion tons/yr of lignocellulose)
  (this is how much electrical power is produced in USA)
- **Salinity Gradient Energy-** Salt & Fresh-waters (global values)
  - **980 GW** (from the 1900 GW available from river/ocean water)
  (20 GW available where WW flows into the ocean)
- **Waste Heat Energy**→ Capture heat in "water" (USA)
  - **500 GW** from industrial "waste heat"
  - **1000 GW** from power plant waste heat
  (Does not include solar and geothermal energy sources)

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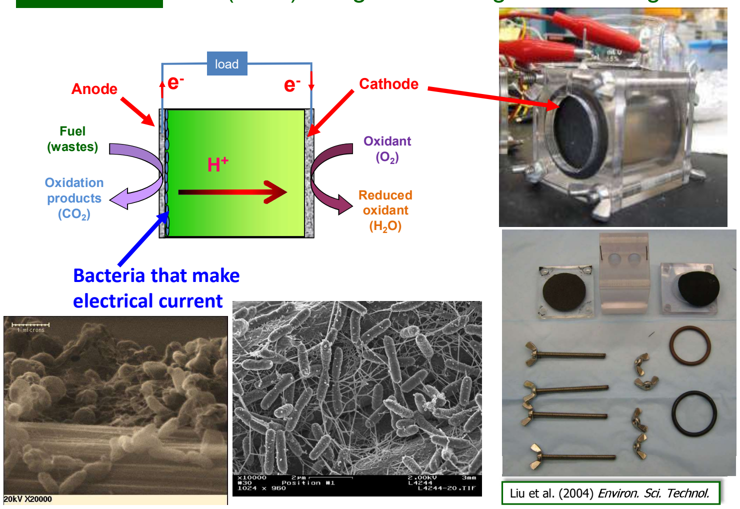
Logan and Rabaey (2012) *Science*
Logan and Elimelech (2012, *Nature*

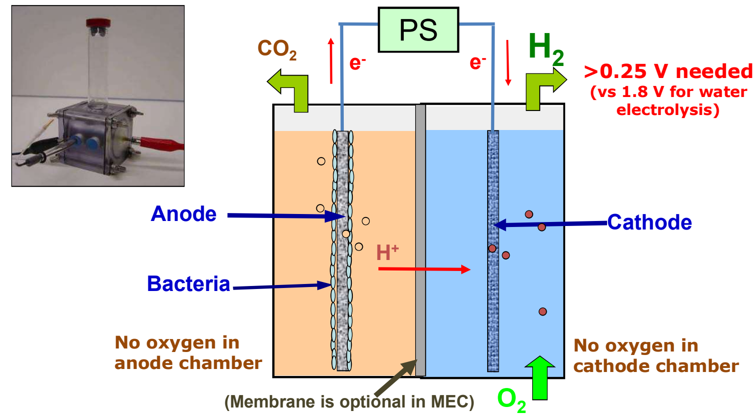
## MFCs Electrical power generation in a **M**icrobial **Fuel** **C**ell (MFC) using exoelectrogenic microorganisms




Liu et al. (2004) *Environ. Sci. Technol.*



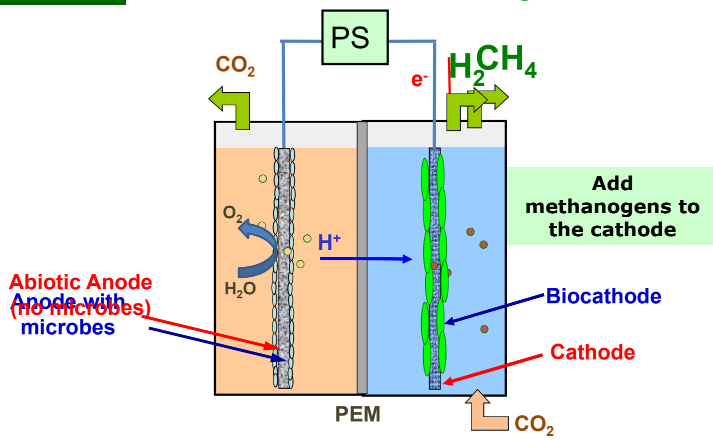
## MECs $H_2$ Production at the cathode using microbes on the anode in **M**icrobial **Electrolysis** **C**ells




Liu, Grot & Logan (2008) *Environ. Sci. Technol.*



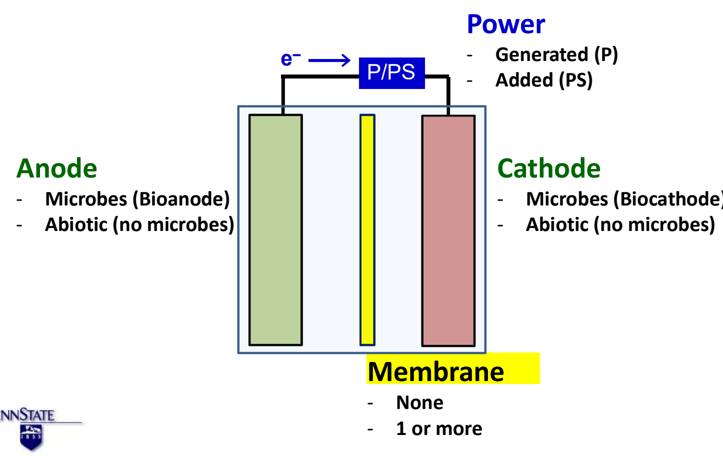
## MMCs $CH_4$ Production at the cathode using microbes on the cathode in **M**icrobial **Methanogenesis** **C**ells



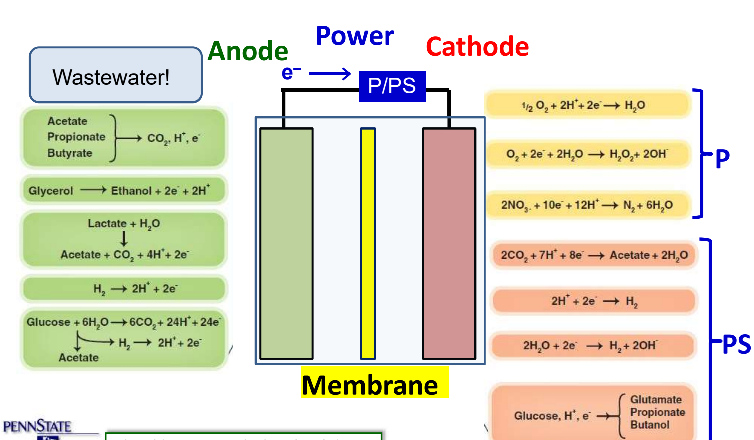

Cheng, Xing, Call & Logan (2009) *Environ. Sci. Technol.*



## MxCs Microbial Electrochemical Technologies (METs)




## METs Microbial Electrochemical Technologies (METs)



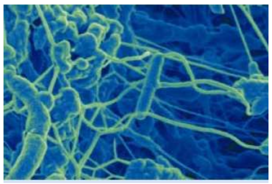

Adapted from: Logan and Rabaey (2012) *Science*

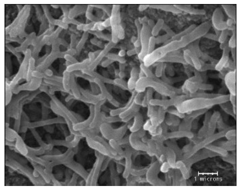
## Focus points

- Electromicrobiology
  - Bioanodes: Electron transfer from bacteria to electrodes
  - (Biocathodes: Biofuel production via electromethanogensis)
- Microbial electrochemical technologies for wastewater treatment
  - Materials
  - Performance
- Scaling up MFCs and MECs
- Conclusions and Acknowledgments

# Electro-active Microorganisms

- Electromicrobiology
  - New sub-discipline of microbiology examining exocellular electron transfer



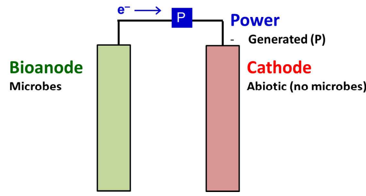




# Electro-active Microorganisms

- Exoelectrogens

  Microbes able to transfer electrons to the outside the cell





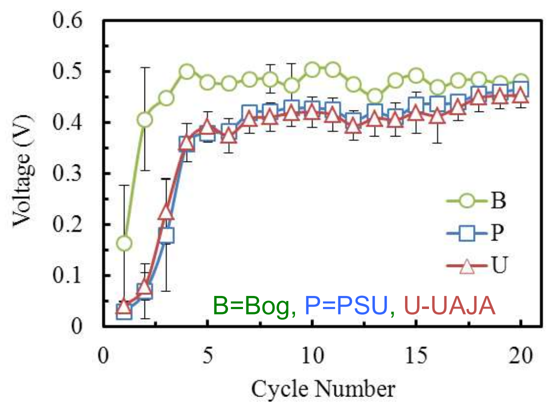

# What microbes are on the anodes?

- Tested reactors over 2 months from 3 sources
  - Penn State wastewater treatment plant (P)
  - UAJA wastewater treatment plant (U)
  - Freshwater bog sediments (B)
- Performance analysis: Power production
- Community analysis
  - Clone libraries
  - Pyrosequencing
  - DGGE
  - FISH

Yates et al. (2012) *ISME J.*



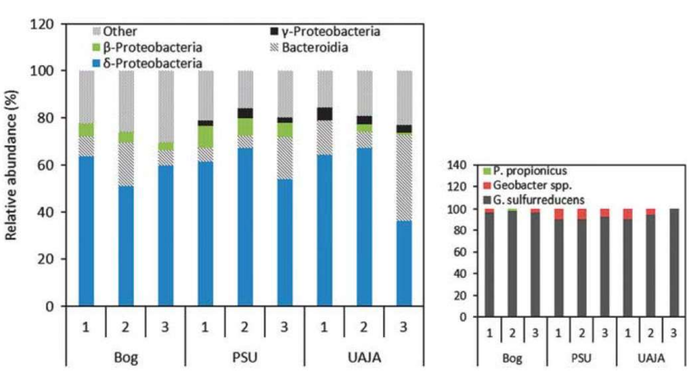
# Bog produced power most rapidly but all inocula converged in power




Yates et al. (2012) *ISME J.*



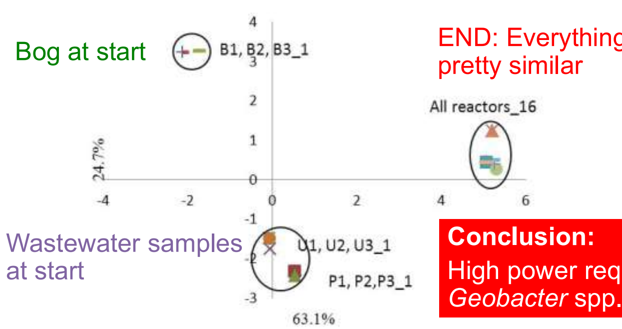
# Pyrosequencing: mostly Delta Proteobacteria… and of those, almost all sequences most similar to *Geobacter sulfurreducens*




Yates et al. (2012) *ISME J.*



# DGGE used to show changes in community diversity over time

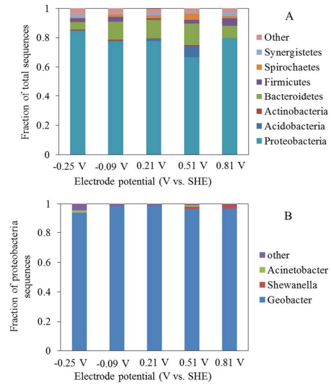



**Conclusion:**
High power requires *Geobacter* spp.

B=Bog, P=PSU, U=UAJA

Yates et al. (2012) *ISME J.*

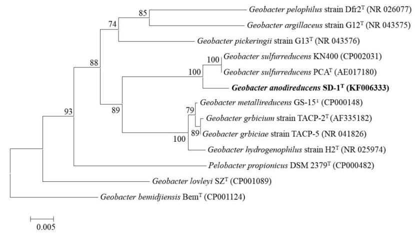
## Community composition unchanged at varied set potentials when different reactors used




Zhu, Yates, Hatzell, Ananda Rao, Saikaly, & Logan (2014) *ES&T*

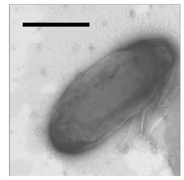


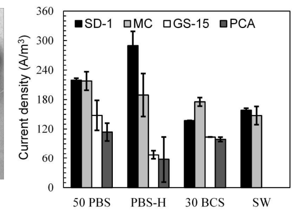
## Isolate from MFC: *Geobacter anodireducens SD-1*



Characteristics of *G. anodireducens* SD-1 (*Geobacter sulfurreducens* PCA)

- Isolated from MFC fed formate, 98% similarity to strain PCA
- Tolerates up to 3% NaCl (vs 1.7% for PCA)
- Grows well in 200 mM phosphate buffer (PCA does not grow)
- Cannot grow using fumarate as electron acceptor (PCA can grow)
- DNA-DNA hybridizations show a G+C content (mol%) of 58.4% (vs 60.9% for PCA)






50 PBS: 50 mM phosphate buffer
PBS-H: 200 mM PBS
30 BCS: 30 mM bicarbonate buffer
SW: 3% NaCl (like seawater)

Sun, Wang, Cheng, Yates, Logan (2014) *Int. J. System. Evol. Microbiol.*

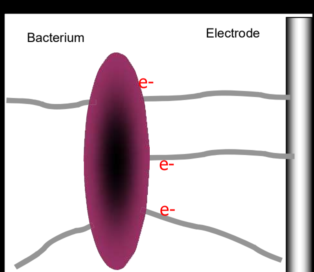
Sun, Call, Wang, Cheng, Logan (2014) *Env. Microbiol. Reports*



## Mechanisms of electron transfer in the biofilm:

Nanowires produced by bacteria !

Gorby & 23 co-authors (2010) *PNAS*



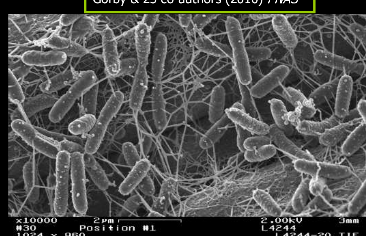
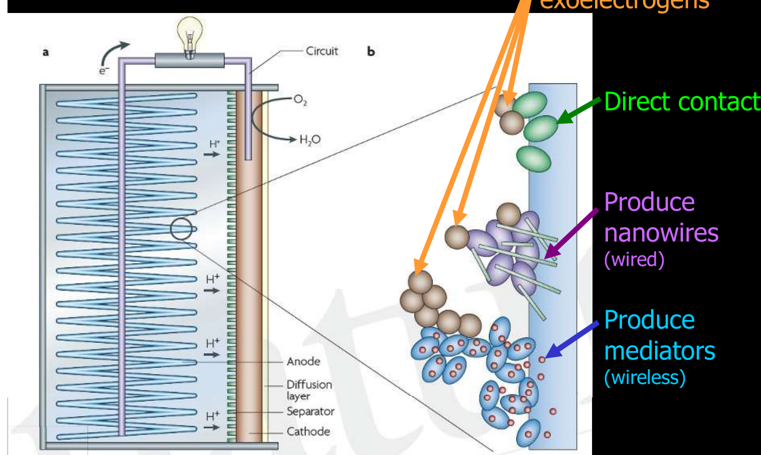



**Figure 1.** Colorized transmission electron micrograph of microbial nanowire networks secreted by *Geobacter sulfurreducens*. Scale bar, 100 nm.

Malvankar & Lovley (2012) *ChemSusChem*



## Electrogenic biofilm ecology

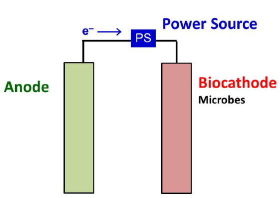



Logan, *Nature Rev. Microbiol.* (2009)



## Electro-active Microorganisms

- Electrotrophs
  Microbes that can accept electrons into the cell

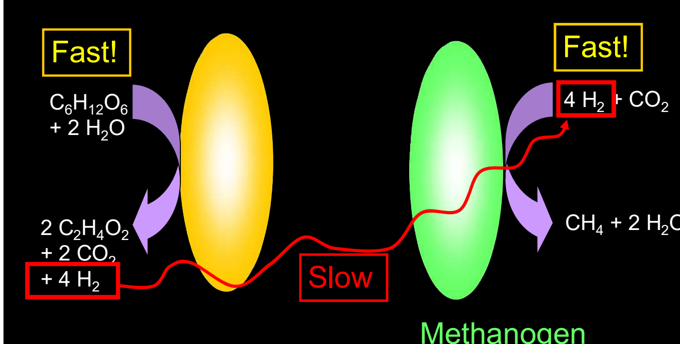



Chemicals used (examples)

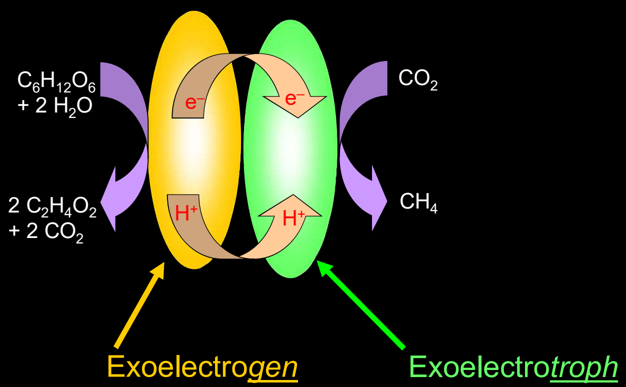
- Dissolved oxygen
- Nitrate
- $CO_2$ - Reduction by methanogens, called "Electromethanogenesis"



## Methanogens: Conventional model based on interspecies *hydrogen* transfer



## New model includes exoelectroactive microorganisms: electron transfer






## What is the evidence for direct electron transfer to methanogens (electromethanogensis)?

- **Identification**: Certain methanogens predominate in mixed culture cathode biofilms
- **Experiments**: Mixed cultures + pure cultures
- **Mechanism**: How are electrons transferred to methanogens?

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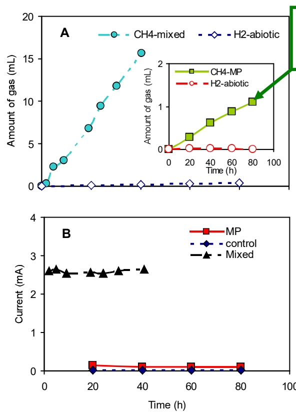
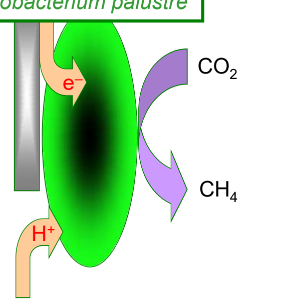
## Electrotrophic Methanogens




Pure culture of ATCC *Methanobacterium palustre*




Mixed culture (*Methanobacterium palustre*)

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Cheng, Call & Logan (2009) *Environ. Sci. Technol.*



## What microbes are on the **cathodes**?

- Tested 2 inoculum sources
  - Anaerobic Digesters (AD), from the Penn State WWTP
  - Freshwater bog sediments (Bog)
- Used different loading rates
  - 0.01% to 25% (mL-sample / mL-medium)
- Performance analysis in 5 mL reactors
  - Methane production
  - Current
- Community analysis: Pyrosequencing

Call & Logan Biosens & Bioelectr 2011

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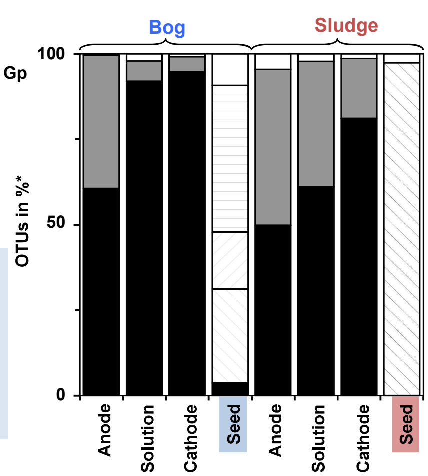
Siegert, Li, Yates, Logan (2015) *Frontiers Microbiol.*



## Reactor Communities (*Archaea*)

- Others <10% each
- Miscellaneous Crenarchaeotic Gp
- Terrestrial Miscellaneous Gp
- *Methanosaeta*
- *Methanobrevibacter*
- *Methanobacterium*




- Final Bog and Sludge communities similar to each other
- Seed very different from final communities
- Mostly *Methanobacterium*

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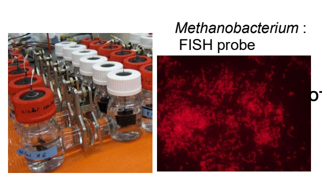
*OTU = operational taxonomic unit

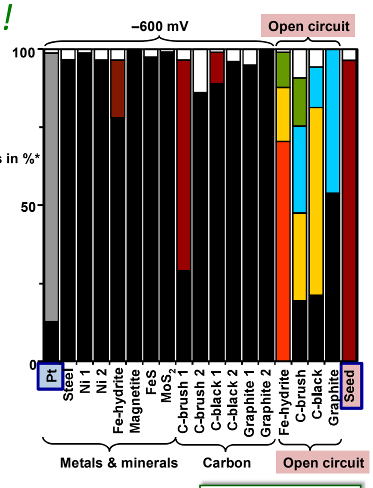
Siegert, Li, Yates, Logan (2015) *Frontiers Microbiol.*



## Which microbe more abundant on different surfaces?

### *Methanobacterium*!

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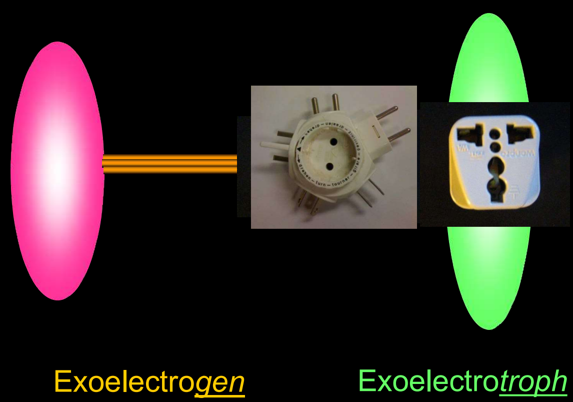
*OTU = operational taxonomic unit

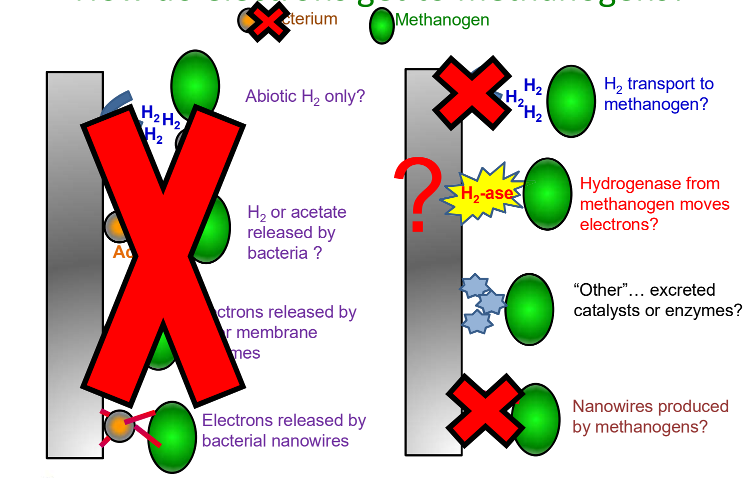
Siegert et al. (unpublished)

# Connections between microbes- Specific or non-specific?






# How do electrons get to methanogens?

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# How do electrotrophic methanogens take in electrons… Are hydrogenases needed?

- Experiments done by Alfred Spormann group at Stanford using:
  - Wild type (**WT**): *Methanococcus maripaludis*, a hydrogenotrophic methanogen (not a *Methanobacterium*)
  - Mutant (**MM1284**): has deletions of 6 hydrogenases (**Δ6**)
    - 5 catabolic hydrogenase genes + 1 anaobolic echB hydrogenase gene
    - grows on methanol, formate, but not $H_2$+$CO_2$
- Two-chamber MECs at set potentials of –600 mV and –700 mV

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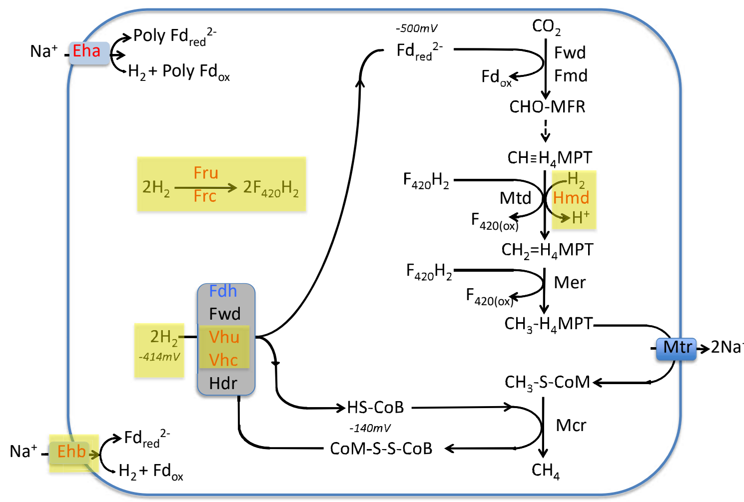
Lohner, Deutzmann, Logan, Leigh, Spormann (2014) *ISME J.*



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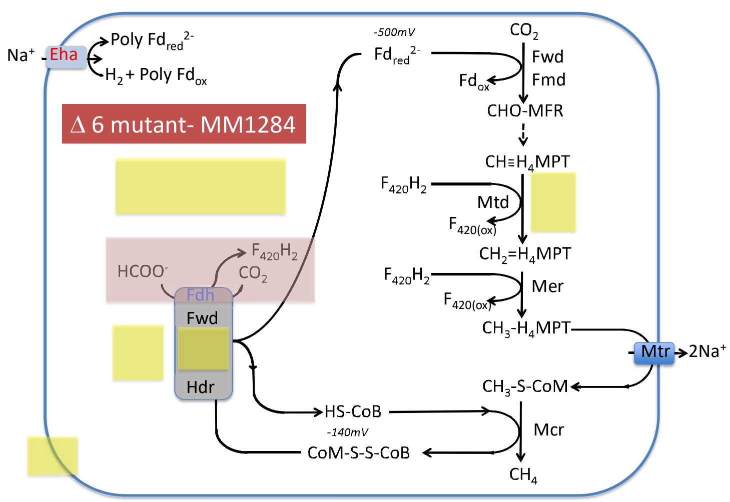
Lohner, Deutzmann, Logan, Leigh, Spormann (2014) *ISME J.*



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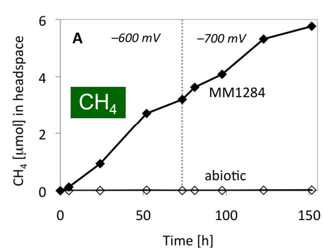
Lohner, Deutzmann, Logan, Leigh, Spormann (2014) *ISME J.*



# Methane production by Mutant MM1284

- Mutant cannot use $H_2$, but produces $CH_4$

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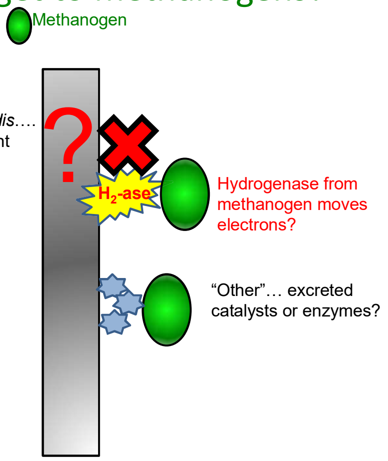
## How do electrons get to methanogens?




So how does *Methanobacterium* make $CH_4$ using electrons derived from the cathode?

… We don't know…

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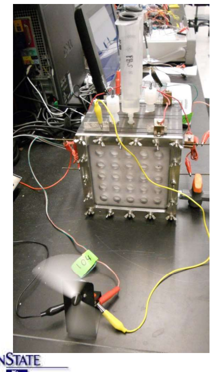
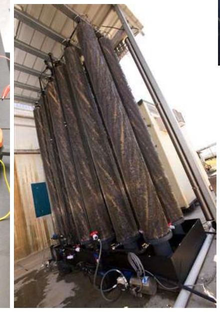
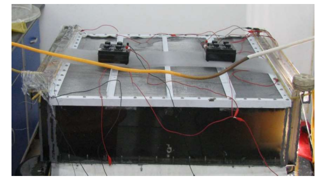
## Scaling up MFCs & MECs

MFCs= fuel cells, make electricity
MECs= electrolysis cells, make $H_2$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## Microbial fuel cells alone cannot be used for wastewater treatment

- MFC Challenges?
  - Reducing material costs
  - Maintaining stable electrogenic populations
- Producing a high quality, treated wastewater needs a secondary treatment process
  - COD removal (solved)
  - Nutrients (in progress)

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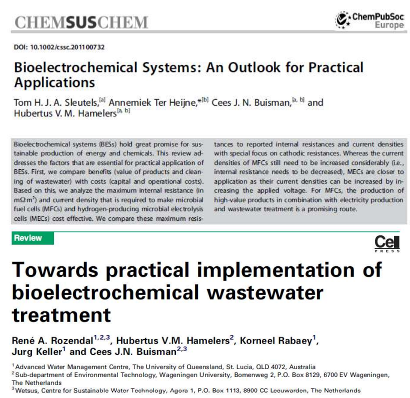
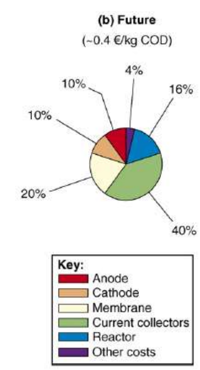
## MFC Architecture


CHEMSUSCHEM
DOI: 10.1002/cssc.201100732
Bioelectrochemical Systems: An Outlook for Practical Applications
Tom H. J. A. Sleutels,[a] Annemiek Ter Heijne,*[b] Cees J. N. Buisman,[a, b] and Hubertus V. M. Hamelers[a, b]

Review
Towards practical implementation of bioelectrochemical wastewater treatment
René A. Rozendal[1,2,3], Hubertus V.M. Hamelers[2], Korneel Rabaey[1], Jurg Keller[1] and Cees J.N. Buisman[2,3]

**Estimates for MFCs**
- 100 € /m² or $130/m²

**Estimates for MECs**
- 100 € /m² or $130/m²

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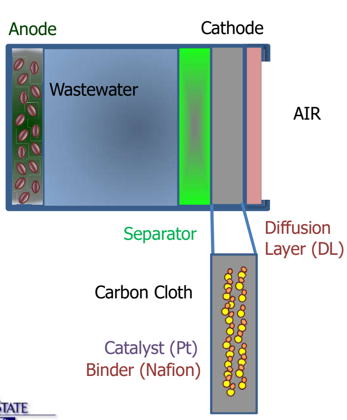
## MFC Architecture




**Original systems: $/m² (US)**
- Carbon cloth~ $1,000
- Pt catalyst~ $ 500
- Binder~ $ 700
- DL (PTFE)~ $ 0.30
- Separator~ $ 1
- **TOTAL $2200**

**New systems: $/m² (US)**
- Anode $20
- Cathode ~~$22~~ **$15**
  - SS + CB= $20
  - Catalyst (AC)=$0.40
  - Binder= $1.5
  - DL (PDMS)= $0.15
- Separator $ 1
- **TOTAL $43**

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## MFC Architecture

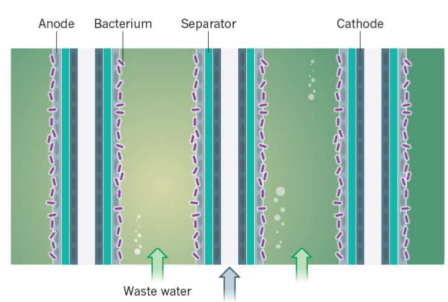


**Figure 3 | An MFC stack.** MFCs are arranged close together to reduce internal resistance and form compact reactors. Within the stack the electrodes consist of repeating units of an anode coated in a mat of bacteria, or biofilm, an insulating separator and a cathode. Waste water flows over the anodes and air over the cathodes. The individual anode and cathode are connected by a wire (not shown).

Logan & Elimelech (2012) *Nature*

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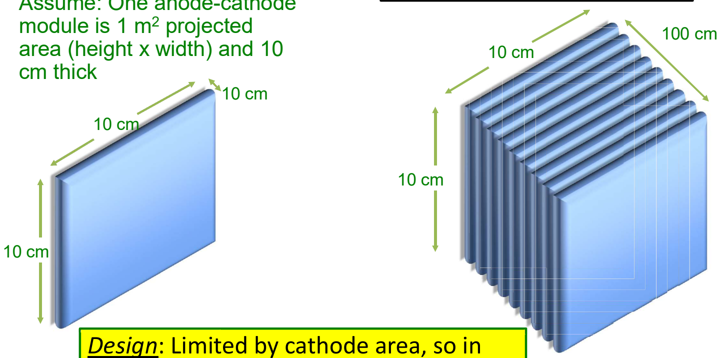
## Overall goal: compact reactor design

Assume: One anode-cathode module is 1 $m^2$ projected area (height x width) and 10 cm thick

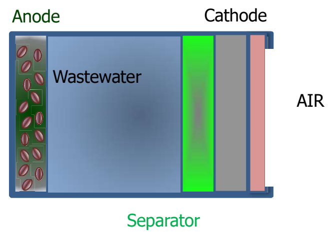
***Result***: 10 modules = 10 $m^2$




***Design***: Limited by cathode area, so in this example we achieve 10 $m^2/m^3$



## MFC Materials




**New systems: $/$m^2$ (US)**

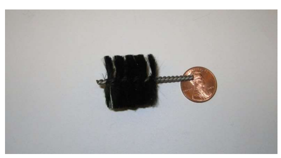
- **Anode** $20
- Separator $ 1
- **Cathode** $15
  - SS mesh
  - AC+Binder
- **TOTAL** **$36**



## MFC Materials

### *Anode*: Graphite brush electrode

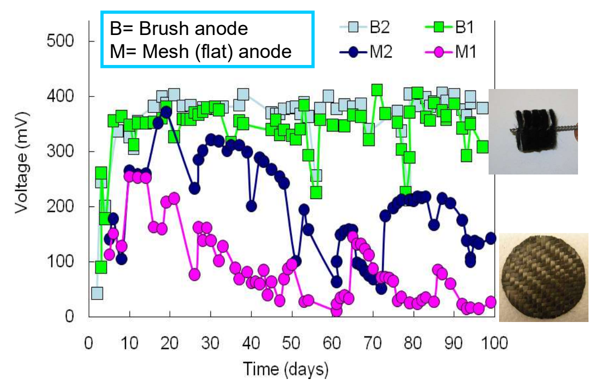
- Graphite fibers commercially available (used in tennis rackets, airplanes, etc.)
- Easy to manufacture
- Fiber diameter- 6-10 µm a good match to bacteria (~1 µm)
- High surface area per volume- Up to 15,000 $m^2/m^3$





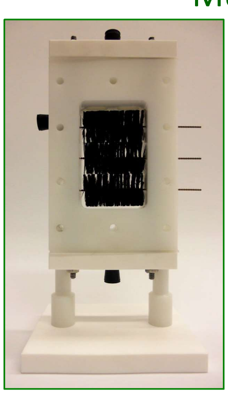
## Voltage Production Results:

Brushes still work better than flat mesh

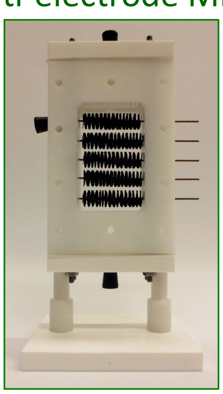




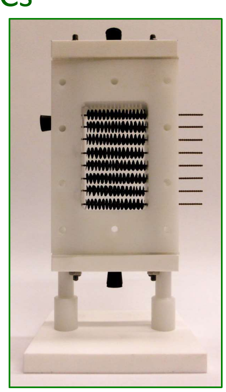

## Multi-electrode MFCs



3 brushes (**R3**)
3500 $m^2/m^3$



5 brushes (**R5**)
2800 $m^2/m^3$



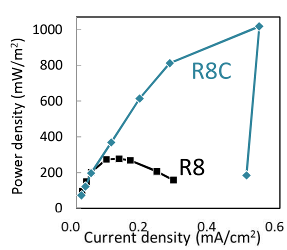
8 brushes (**R8**)
2900 $m^2/m^3$

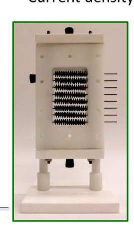
Electrode area (2.5 cm diameter brush/chamber width = 40 $m^2/m^3$



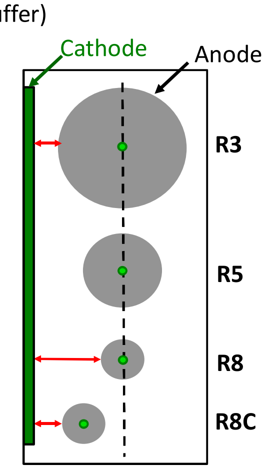
## Smaller, closer brushes work best

(Continuous flow, acetate in buffer)

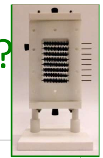


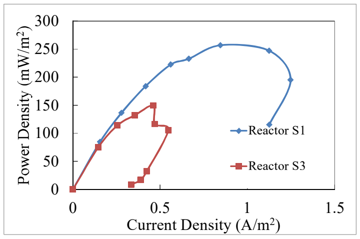
Maximum power densities
R8C= 1020 mW/$m^2$
R8= 280 mW/$m^2$
(R3= 560 mW/$m^2$)
(not shown)

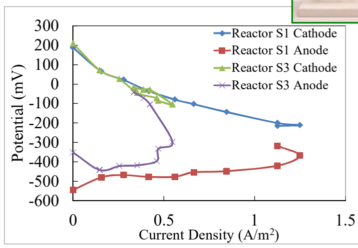


## Reactor stability with smaller brushes?



Continuous flow, 4 hr HRT, domestic ww




Maximum power quite different:
**260 mW/m²** vs **150 mW/m²**
Not possible to get true "duplicates"

- Cathode performance similar
- Anodes performance unstable (S3)

**Conclusion:** Thin brushes led to conditions similar to "flat anodes", where $O_2$ transfer through the cathode affected anode performance

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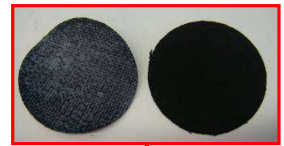
Stager & Logan, *Unpublished*

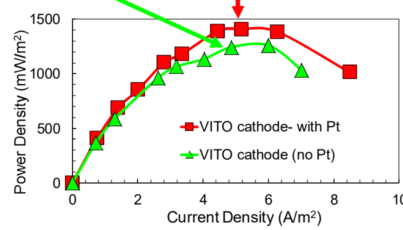
## Cathode: Activated Carbon Catalysts

VITO cathode (no Pt)

Carbon cloth with Pt



Activated carbon cathode works almost as well as Pt catalyst




PENNSTATE vito vision on technology

Zhang, Cheng, Van Bogaert, Pant & Logan (2009) *Electrochem. Commun.*



## Catalytic Activity of ACs

| Precursor | Sample |
|---|---|
| Hardwood | **W1-** MWV1500 |
| Phenol resin | **R1-** Kuraray RP-20 |
| Peat | **P1-** Norit SX1 |
| Peat | **P2-** Norit SX Plus |
| Peat | **P3-** Norit SX Ultra |
| Coconut shell | **C1-** Kuraray YP-50 |
| Coconut shell | **C2-** CR8325C |
| Coconut shell | **C3-** ACP1250 |
| Bituminous Coal | **B1-** CR325B |

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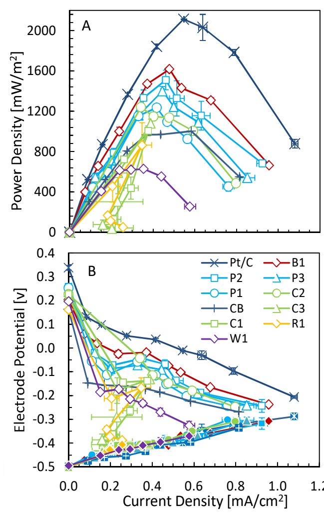
Watson & Logan (2013) ES&T



## Performance in MFCs




- Pt is still the best
- B1 little better than P2
  - LSV: P2 > B1
  - Differences due to cathode construction as AC added to rotating disc electrode

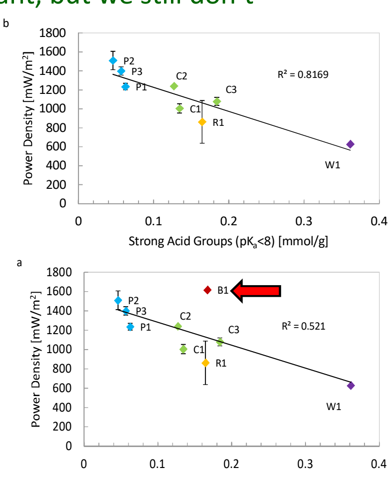
| Precursor | Sample |
|---|---|
| Hardwood | **W1-** MWV1500 |
| Phenol resin | **R1-** Kuraray RP-20 |
| Peat | **P1-** Norit SX1 |
| Peat | **P2-** Norit SX Plus |
| Peat | **P3-** Norit SX Ultra |
| Coconut shell | **C1-** Kuraray YP-50 |
| Coconut shell | **C2-** CR8325C |
| Coconut shell | **C3-** ACP1250 |
| Bituminous Coal | **B1-** CR325B |

Watson & Logan (2013) ES&T



## Carbon chemistry important, but we still don't understand it.

- Carbon titrated to determine relative abundance of strong acid functional groups
- Correlation "significant" only if B1 (bituminous coal) is exluded…
  - B1 worked well as an oxygen reduction catalyst, so other factors important
  - Possible?: Pore size, surface area, other surface chemical characteristics.

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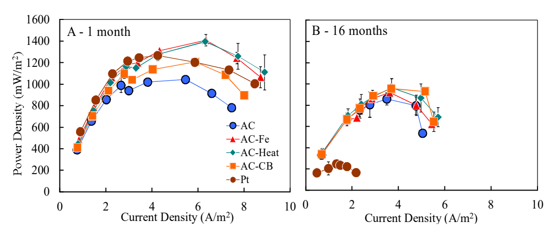
Watson, Delgado & Logan (2013) ES&T



## Activated Carbon Cathodes- (Manufactured by VITO)

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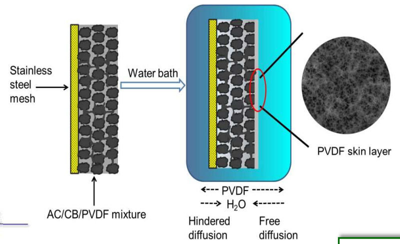
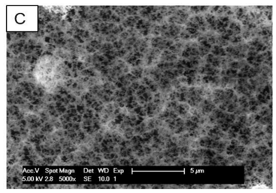
Zhang, Pant, Zhang, Liu, Logan (2014) *Chem. Electro. Chem.*

## New Binder for Activated Carbon: PVDF

- Using a PVDF binder is simple and effective
  - 1- apply to SS mesh; 2- phase inversion in water
  - Make at room temperature
  - Amenable to continuous rolling process
  - No separate gas diffusion layer (GDL) needed
  - Cost: \$15 $m^{-2}$ (\$12 $m^{-2}$ for SS mesh, \$3 $m^{-2}$ for catalyst and binder)



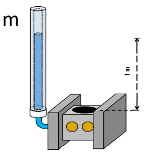



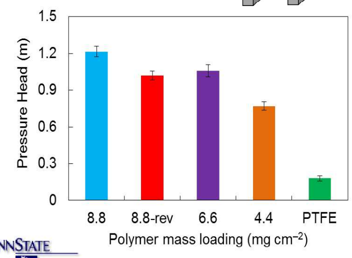

Yang, He, Zhang, Hickner, Logan (2014) *ES&T Letters*



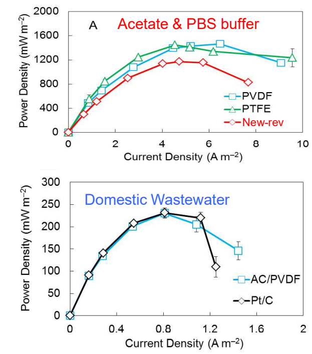
## PVDF Binder

- Water pressure up to 1.2 m (vs 0.2 m for PTFE)






Power the same as PTFE applied to carbon cloth/Pt



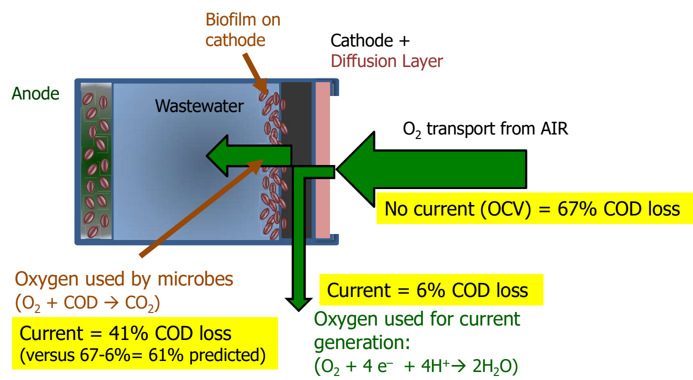

Yang, He, Zhang, Hickner, Logan (2014) *ES&T Letters*



# MFCs and MECs for Wastewater Treatment

## …and why MxCs alone cannot accomplish wastewater treatment

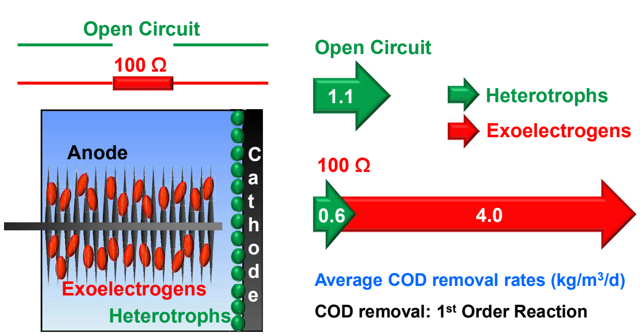
## Oxygen used for current generation decreases $O_2$ crossover, increases Coulombic efficiency… a little…




Ren, Zhang, He, Logan (2014) *Biotechnol. Bioeng.*



## Curreng generation shifts more substrate to electricity generation in MFCs (acetate)




Zhang, He, Ren, Evans, Logan (2015) *Biores. Technol.*

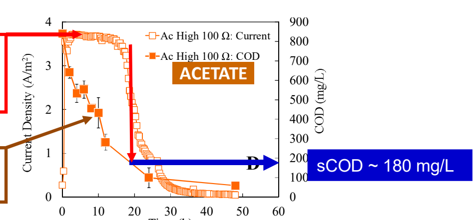


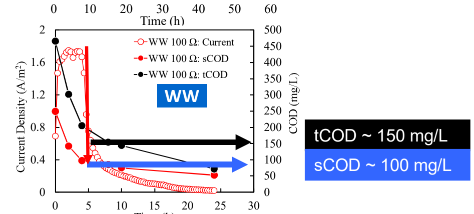
## Low sCOD limits current generation!

Current very stable, then it suddenly decreases

COD removal constant (first-order kinetics)

In both cases, COD continues to be removed, so treatment continues without electricity generation, but ww cannot be discharged due to high COD

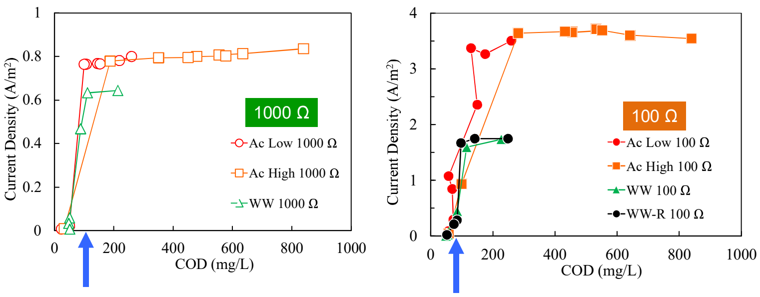



Zhang, He, Ren, Evans, Logan (2015) *Biores. Technol.*

# Current density vs soluble COD (sCOD)

Current rapidly drops off at ~100 mg/L sCOD




In both cases, current rapidly decreases when sCOD is still high (~100 mg/L)

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Zhang, He, Ren, Evans, Logan (2015) *Biores. Technol.*



# Reason(s) for rapid decline in current?

- High sCOD: Chemical flux (J) into the biofilm is first order (consistent with a "fast reaction")

$k_1$= first order rate constant (function of microbial kinetics)

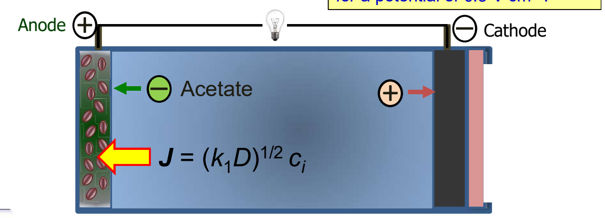
$D$ = Diffusion constant

→ But *D is a function of the electric field* $\nabla\varphi$ (Nernst-Plank eqn.)

$$J = [uc_i - D\nabla c_i] - \frac{z_i DF\nabla\varphi}{RT}c_i = [0 - D\nabla c_i] - \frac{z_i DF\Delta E}{RT\,\Delta x}c_i$$

$$= -\left(1+\frac{F\nabla E}{RT}\right)D_{eff}\frac{\Delta c_i}{\Delta x}$$

Term is red increases $D$ by 11x for a potential of 0.3 V cm⁻¹.

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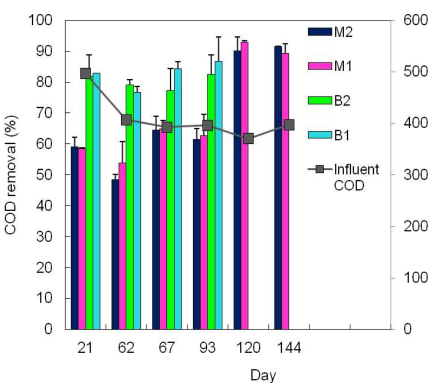
# How much COD removal can we get from domestic WW?
## ~80-90%, but final COD is too high




- Influent

COD = 400 mg/L or $BOD_5$ = 200 mg/L

- Effluent

$BOD_5$ 35-40 mg/L

(like a Trickling Filter)

Removal >80%

- Why isn't more COD removed?
- What is "fate" of COD?

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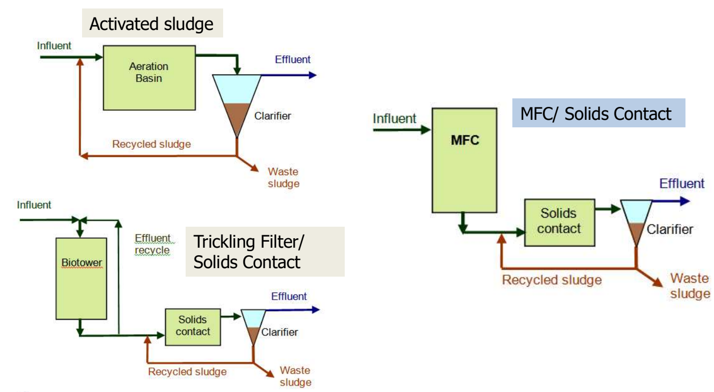
Hays and Logan (2011) *J. Power Sources*



# Solids Contact Process Improves Performance of Fixed Film Bioreactors

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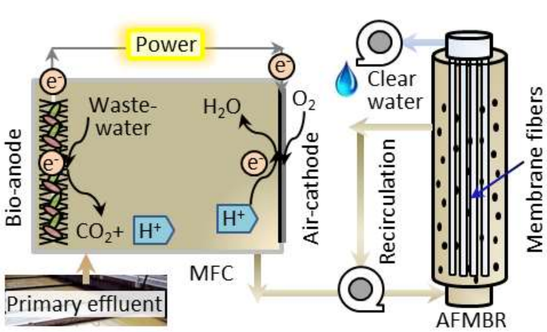
Logan (2008) *"Microbial Fuel Cells"*



# MFC + AFMBR
## (Anaerobic Fluidized Bed Membrane Bioreactor)

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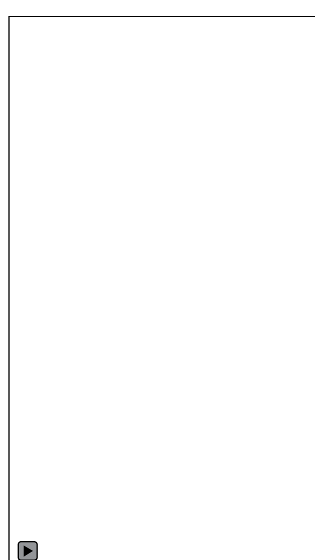
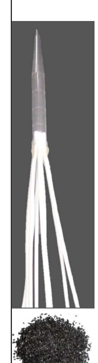
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*





# AFMBR Construction

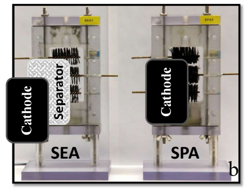


- Idea of AFMBR first published by Chae et al. (ES&T). Used as a second stage to granular fluidized bed anerobic digester
- AFMBR consists of a reactor body + ultrafiltration membrane + granular activated carbon (GAC)
- GAC fluidized by recirculation
- In tests here, used with a hydraulic retention time of 1 hour

Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*

# Generation I: MFC configuration

- SEA: Separator electrode assembly
  - Trimmed graphite fiber brush, one side flat
  - Separator between brush anode and cathode placed together
- SPA: Spaced electrode assembly
  - Brush placed distant from cathode so it can't touch it
- Two reactors used in series
  - 2 × 4 h HRT = 8 h HRT
- Total of 4 MFCs (2 SEA, 2 SPA)

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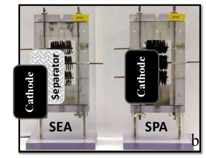
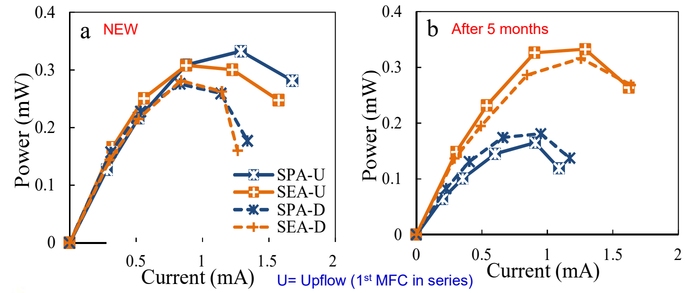
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*



# Comparison of performance of SEA and SPA MFCs over time

- Initially: similar performance
- After 5 months: SEA>> SPA




U= Upflow (1st MFC in series)
D=downflow (2nd MFC in series)

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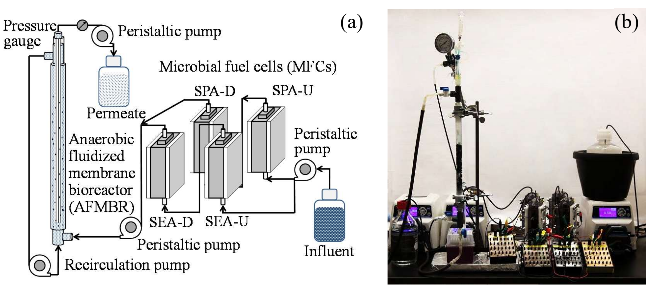
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*



# Experimental Setup: MFC+AFMBR




Reactor HRTs: MFC=4 h (each); AFMBR=1 h
"F"= Granular activated carbon (GAC), fluidized, used for biofilm support and membrane cleaning (scour)
"MBR": PVDF hollow fiber membranes
MFC types: SEA (separator); SPA (spaced, no separator)

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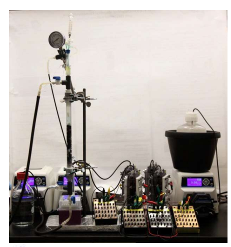
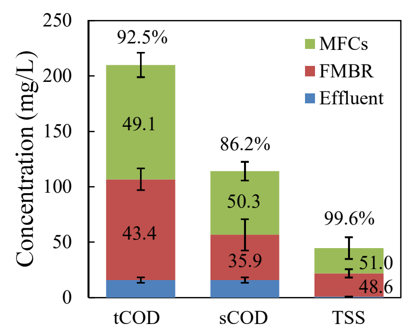
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*



# Effluent reduced to 16 mg/L tCOD

- Two trains of MFC (HRT = 4 h) to AFMBR (HRT = 1 h)
- Membrane flux 16 L/m$^2$/h
- 50 days performance
- Energy balanced
  (MFC produced = AFMBR used)






WW in FMBR    Permeate

- Effluent COD = 16 mg/L
- Effluent TSS <1 mg/L

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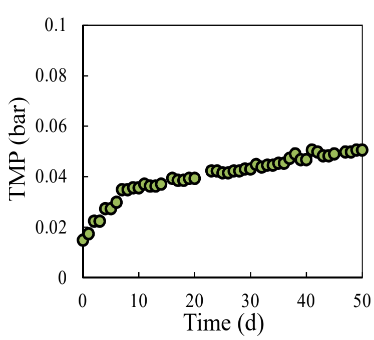
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*



# Key to AFMBR success: Little fouling

- First 10 days, initial rapid increase in transmembrane pressure (TMP)
- Days 10-50, only slight increase in TMP
- Flux of 16 LMH much greater than that in previous studies with anaerobic fluidized bed reactors (AFBRs)
  - AFMBR: 6-7 LMH at start
  - 4-11 LMH at start

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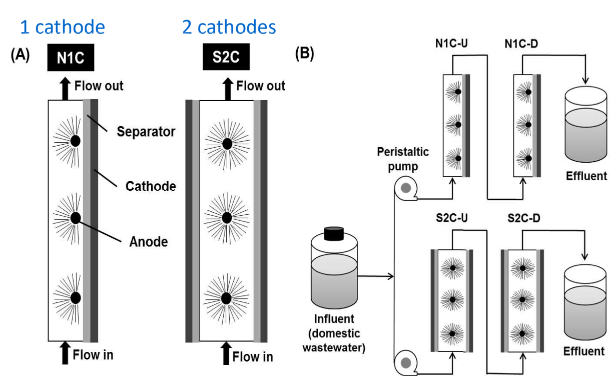
Ren, Ahn & Logan (2014) *Environ. Sci. Technol.*



# Gen I MFCs: Separators & 1 or 2 Cathodes




N1C=SEA (previous terminology)

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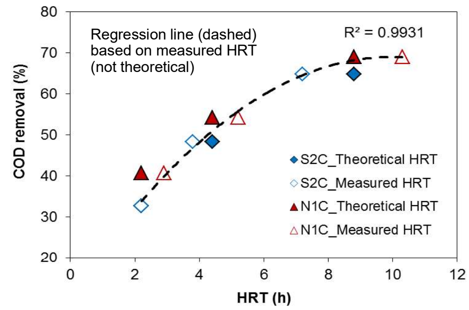
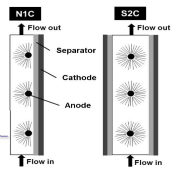
Kim, Yang and Logan (2015) *In preparation*

# COD Removals corrected for Actual HRTs

## HRTs: N1C > S2C

- Same theoretical HRT set for reactor comparisons
- N1C > S2C
- It is important to *measure* actual HRTs

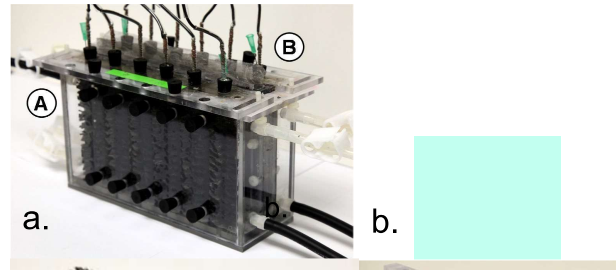



Kim, Yang and Logan (2015) *In preparation*



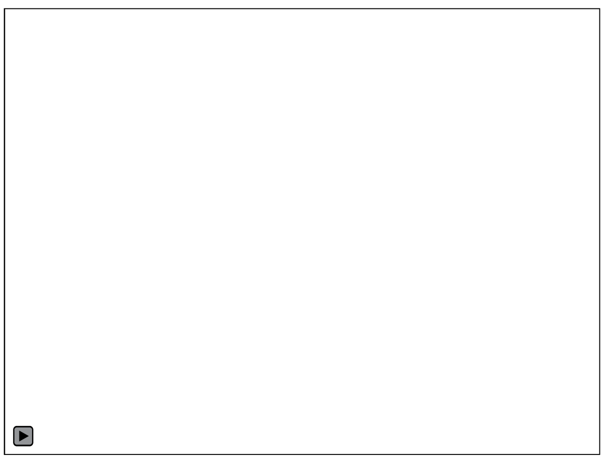
# Generation II (Gen II) MFCs



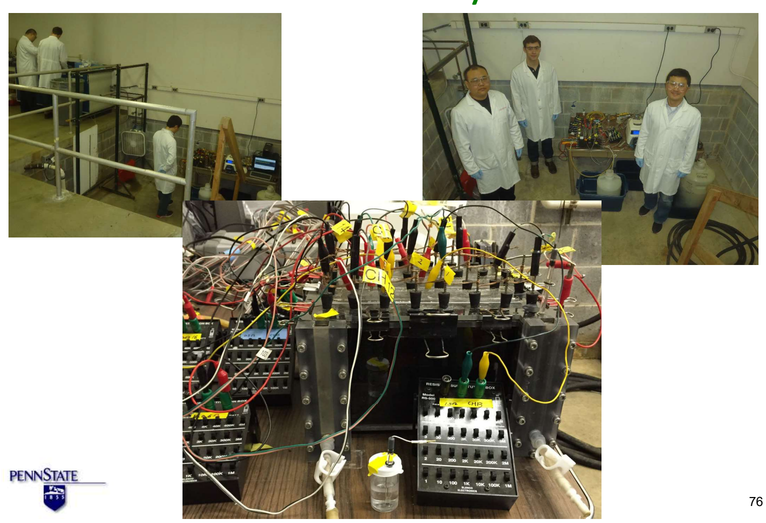

- Modular MFC
  - Shown with 2 anodes, 2 cathodes
  - Produced ~ 400 mW/m$^2$ with domestic wastewater

He, Zhang, Logan (2014) *In preparation*





# Gen III MFCs: Stay tuned!





# Conclusions

- New renewable energy technologies can be created using electro-active microorganisms:
  - Exoelectrogens- make electrical current
  - Electrotrophs- consume electrons, make $H_2$ and $CH_4$
- MFCs for wastewater treatment
  - Cost: Reduced to < $40 m$^{-2}$
  - Materials: Brush anodes; separator; Activated Carbon cathodes with stainless steel mesh current collectors, PVDF binder
  - Power: Don't try to fully treat wastewater; remove COD to about 100-150 mg/L
- + AFMBR
  - Add a secondary treatment system to remove COD to <20 mg/L
  - TSS < 1 mg/L, so no secondary clarifier needed



# Thanks to students and researchers in the MxC team at Penn State!

Current research sponsors
SERDP/DOD (2012-2015); GCEP/Stanford (2012-2015); NREL/DOE (2014-2017); NSF SusChem-EAGER (2015-2016)

# International Collaborations