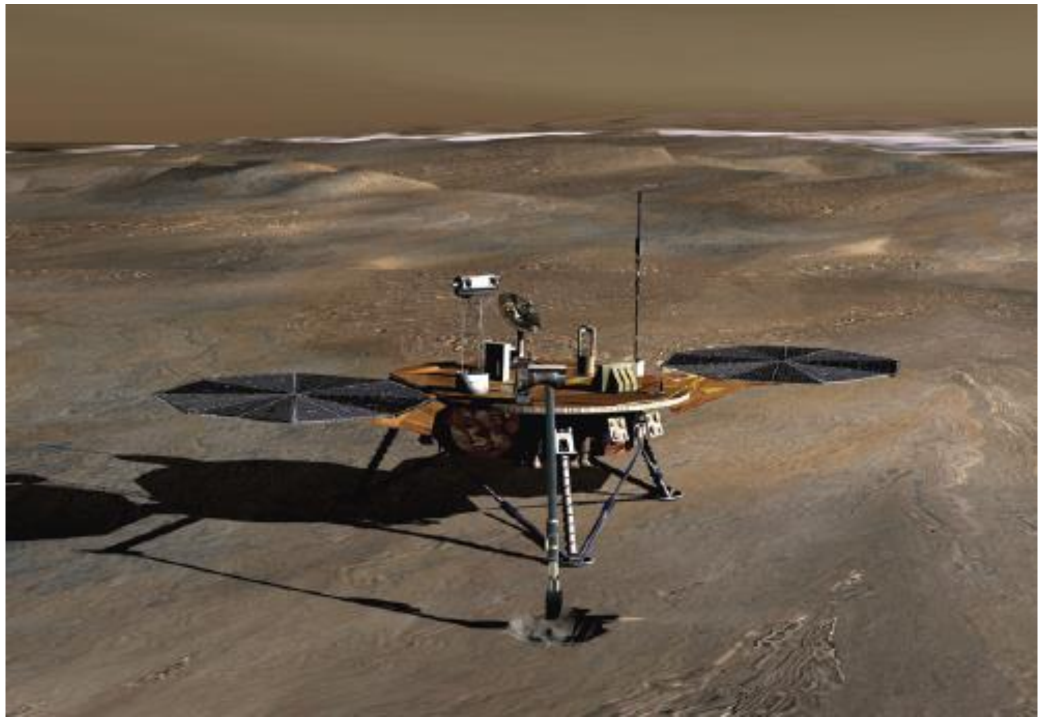
Physics-Based Modeling and Simulation of Emerging Battery Technologies for **Aerospace**

Mohit Mehta and John Lawson

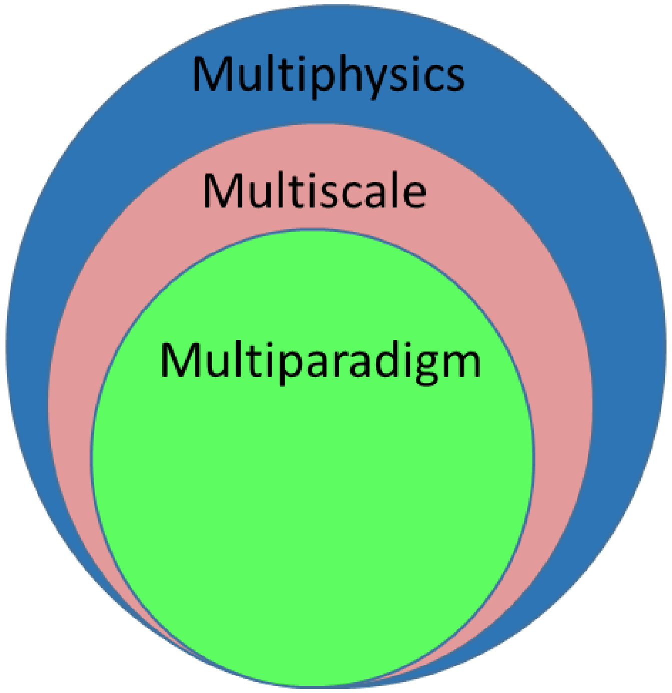
Presentation date: 08/21/2019

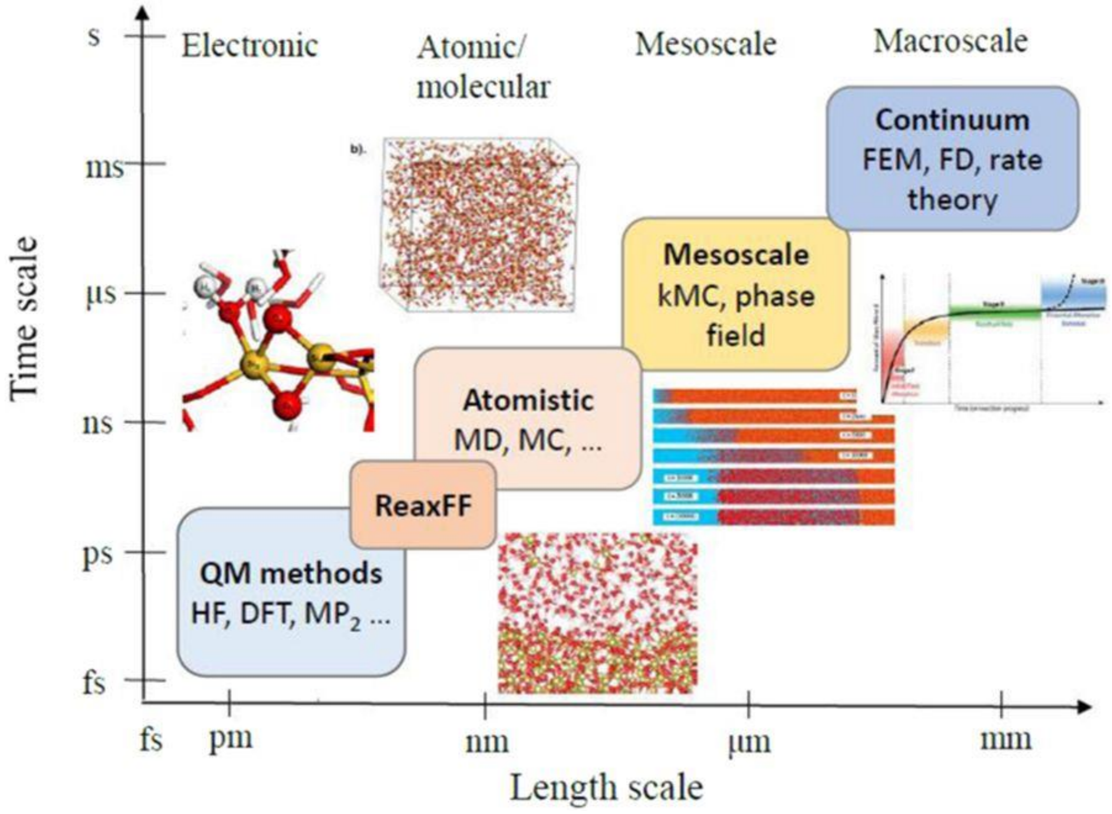
1. Batteries for Aviation
2. Batteries for Space



Courtesy NASA/JPL-Caltech

Introduction to modeling and simulation

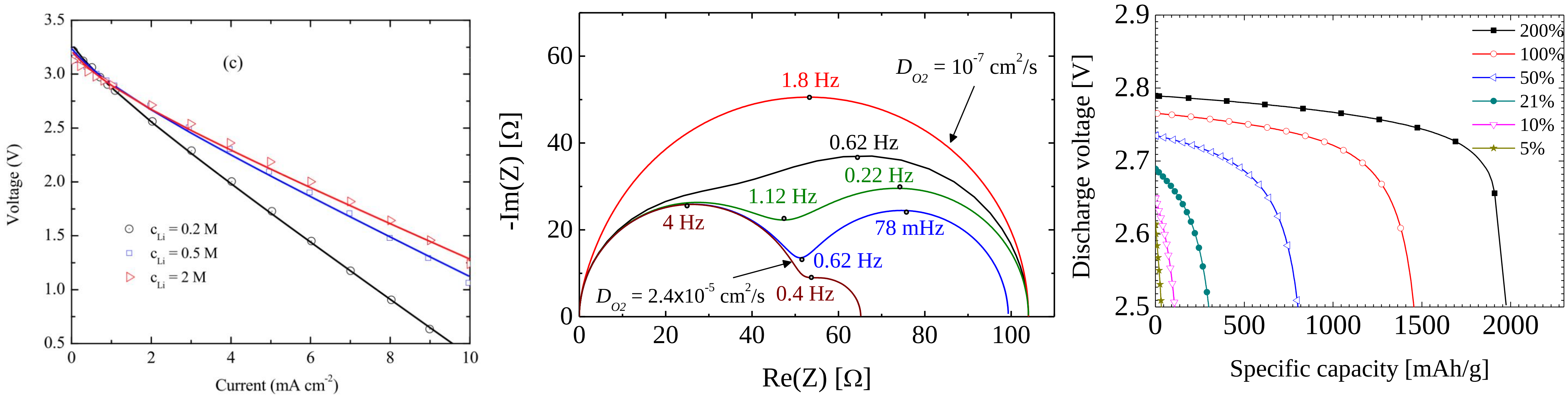




Du, J. & Rimsza, J. M. Atomistic computer simulations of water interactions and dissolution of inorganic glasses. *npj Mater Degrad* **1**, 1–12 (2017).

Simulation

Experiments

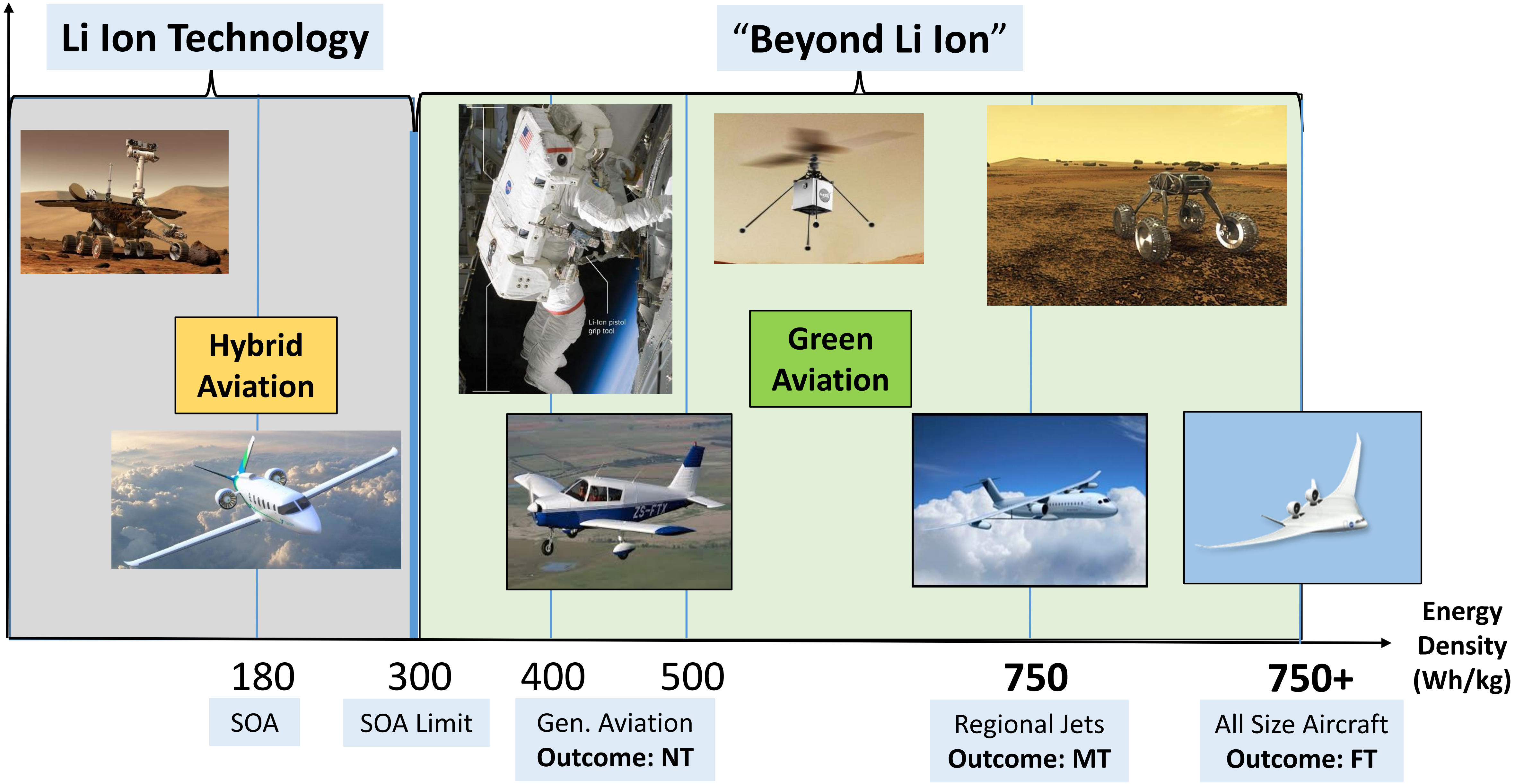


Mehta, M. & Andrei, P., 2014. *ECS Transactions*, 61(15), pp.39–55

Aerospace Battery Requirements

Major requirement is: High Energy Density

Other requirements are **rechargeable**, **safety**, power, recharge time, cost, etc.



Li Ion Technology

"Beyond Li Ion"

Hybrid Aviation

Green Aviation

Energy Density (Wh/kg)

180
SOA

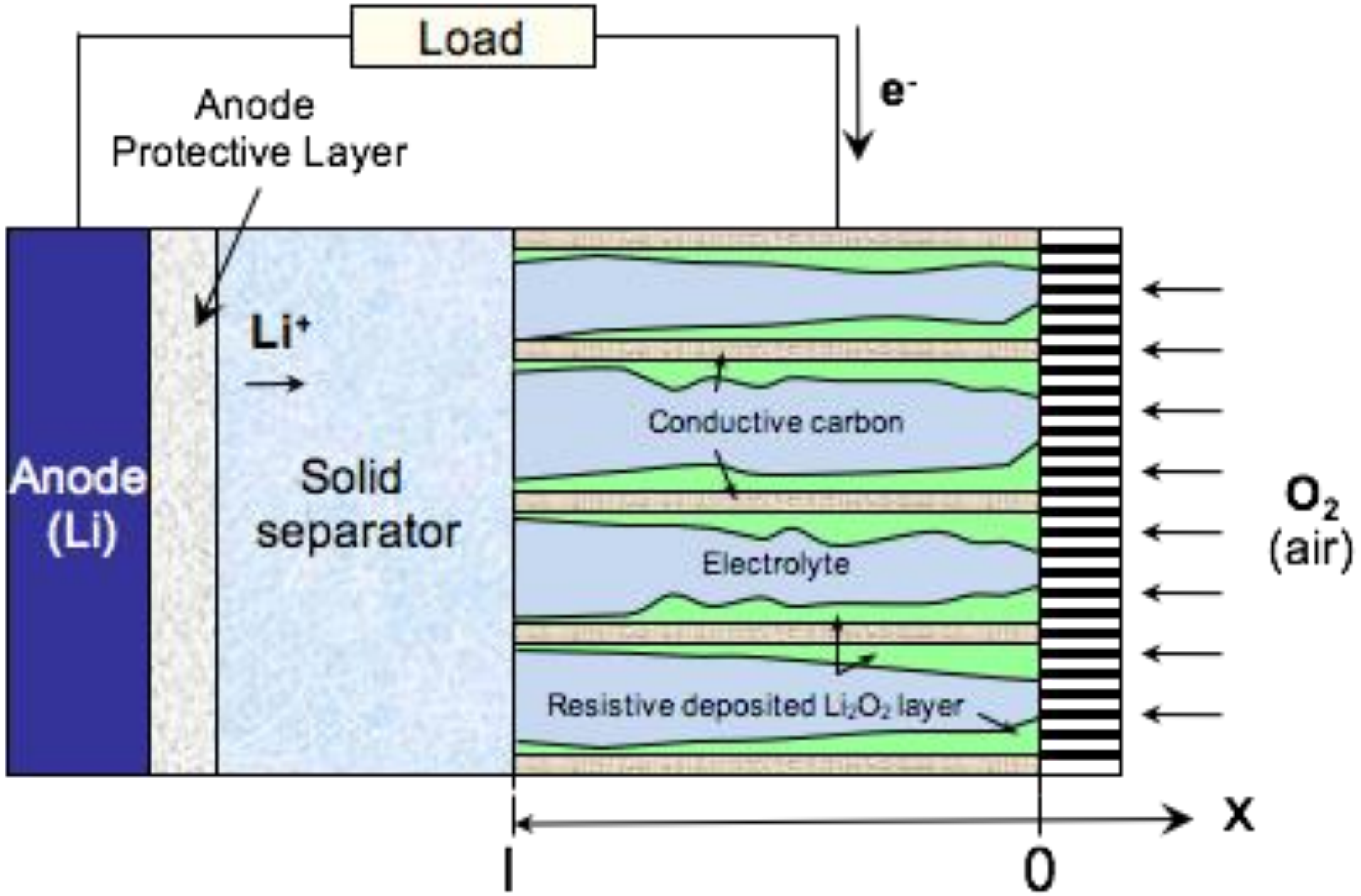
300
SOA Limit

400 500
Gen. Aviation
Outcome: NT

750
Regional Jets
Outcome: MT

750+
All Size Aircraft
Outcome: FT

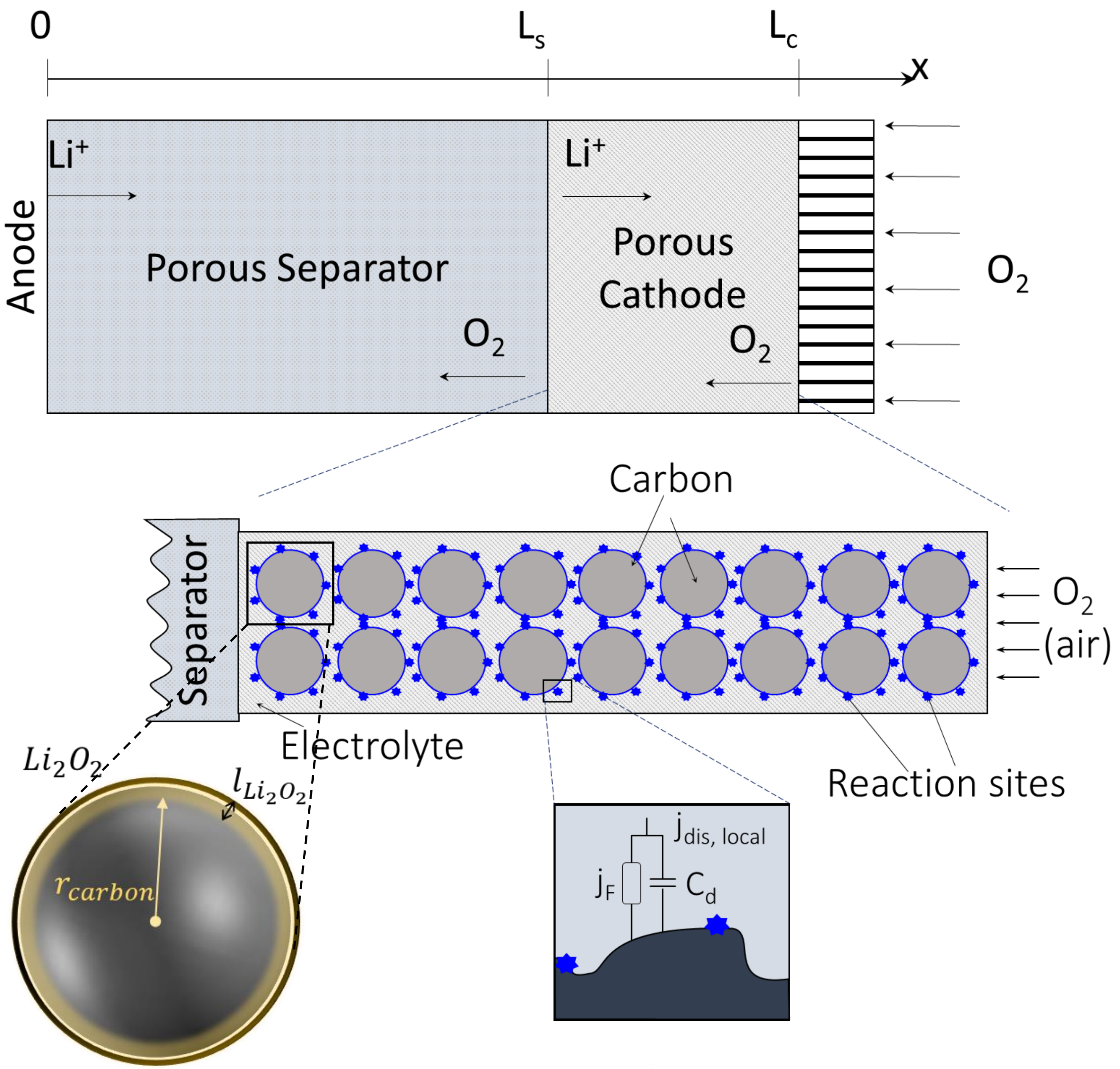
Batteries for Aviation (exploring Li-O_2)



Discharge:	$\mathrm{Li} \rightarrow \mathrm{Li}^+ + e^-$	$2\mathrm{Li}^+ + \mathrm{O}_2 + 2e^- \rightarrow \mathrm{Li}_2\mathrm{O}_{2(s)} \downarrow$
Charge:	$\mathrm{Li}^+ + e^- \rightarrow \mathrm{Li} \downarrow$	$\mathrm{Li}_2\mathrm{O}_{2(s)} \rightarrow 2\mathrm{Li}^+ + 2e^- + \mathrm{O}_2 \uparrow$

Mehta, M. & Andrei, P., 2014. *ECS Transactions*, 61(15), pp.39–55

Modeling a lithium-oxygen battery



$$V_{\text{discharge}} = j_{\text{dis}}\delta_{Li_2O_2}\rho_{\text{Li}_2\text{O}_2}\exp\left(\alpha_{j_{dis}}\frac{-\delta_{Li_2O_2}}{10\text{nm}}\right)$$

Reaction rate
$$R_C = nFc_{\text{O}_2}k_0a\left(e^{\frac{(1-\beta)n}{V_T}\eta} - e^{\frac{-n\beta}{V_T}\eta}\right)$$

Oxygen dissolvation
$$c_{O_2}(L_c) = N_{O2} = k_f[p_{O2} - k_Hc_{O2}(0)]$$

$$\overbrace{\eta}^{\text{Over-voltage}} = \underbrace{\phi_{\text{Li}}}_{\text{electrolyte}} - \underbrace{\phi}_{\text{electrode}} - \overbrace{E^0}^{\text{thermodynamic}} - \underbrace{V_{\text{discharge}}}_{\text{Li}_2\text{O}_2}$$

$-I$ (electron current)

$$\nabla\cdot\left(\overbrace{\sigma_{\text{eff}}\nabla\phi}\right) + R_{\text{C}} = aC_d\frac{\partial(\phi - \phi_{\text{Li}})}{\partial t}$$

$-I_{\text{Li}}$ (electrolyte current)

$$\nabla\cdot\left(\overbrace{\kappa_{\text{eff}}\nabla\phi_{\text{Li}} + \kappa_{\text{D}}\nabla\ln c_{\text{Li}}}\right) - R_{\text{C}} = aC_d\frac{\partial(\phi - \phi_{\text{Li}})}{\partial t}$$

$-I_{\text{Li}}$ (electrolyte diffusion flux)

$$\frac{\partial(\epsilon c_{\text{Li}})}{\partial t} = \nabla\cdot(\overbrace{D_{\text{Li,eff}}\nabla c_{\text{Li}}}) - \frac{1-t^+}{F}R_{\text{C}} - \frac{I_{\text{Li}}\cdot\nabla t^+}{F}$$

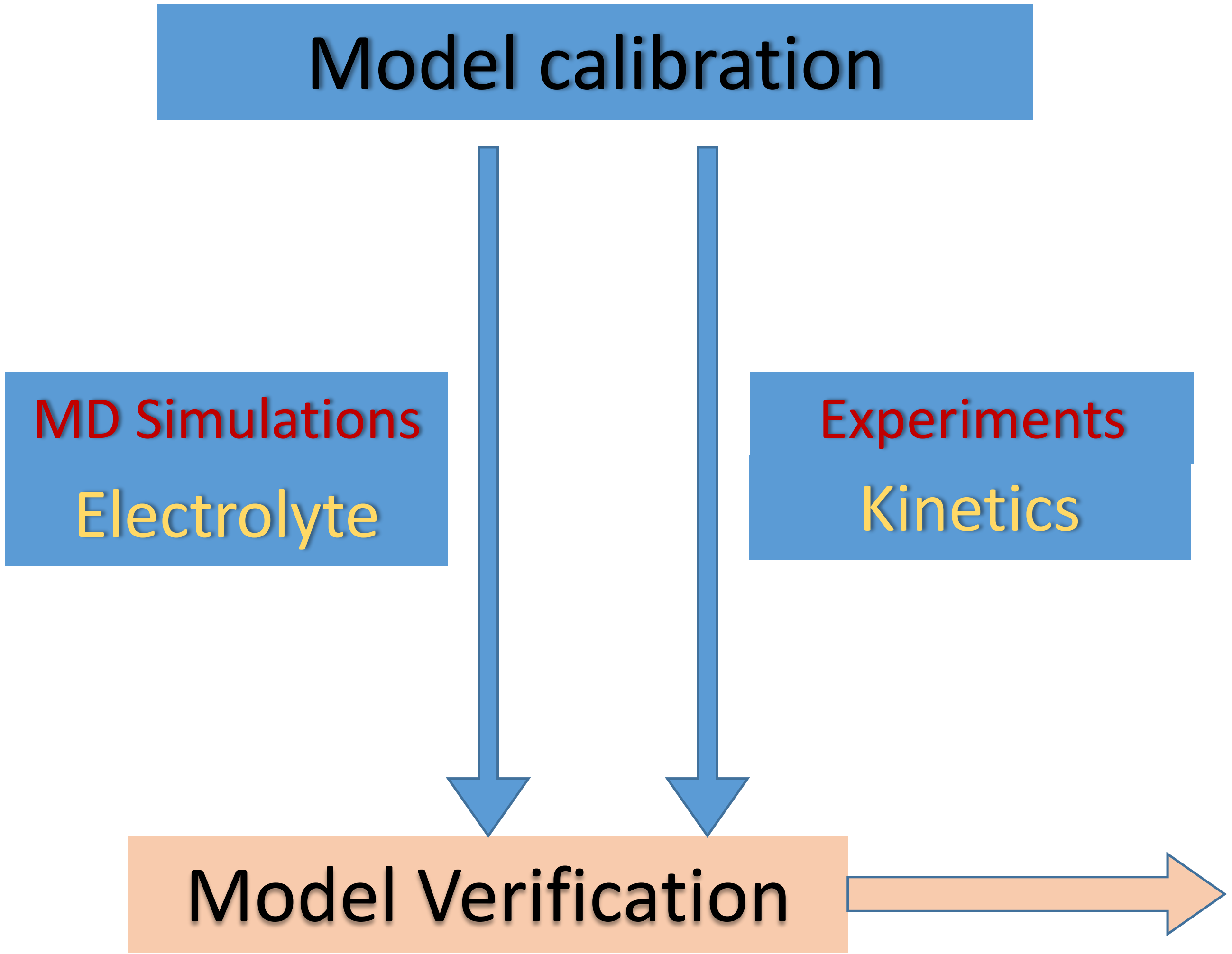
$-I_{\text{O2}}$ (O$_2$ diffusion flux)

$$\frac{\partial(\epsilon c_{\text{O}_2})}{\partial t} = \nabla\cdot(\overbrace{D_{\text{O}_2,\text{eff}}\nabla c_{\text{O}_2}}) - \frac{R_C}{nF}$$

ϵ (porosity change -from Li$_2$O$_2$ deposition)

$$\overbrace{\frac{\partial\epsilon}{\partial t}} = -R_C\frac{M_{\text{discharge}}}{nF\rho_{\text{m,discharge}}}$$

Model calibration for simulating high current



Model calibration

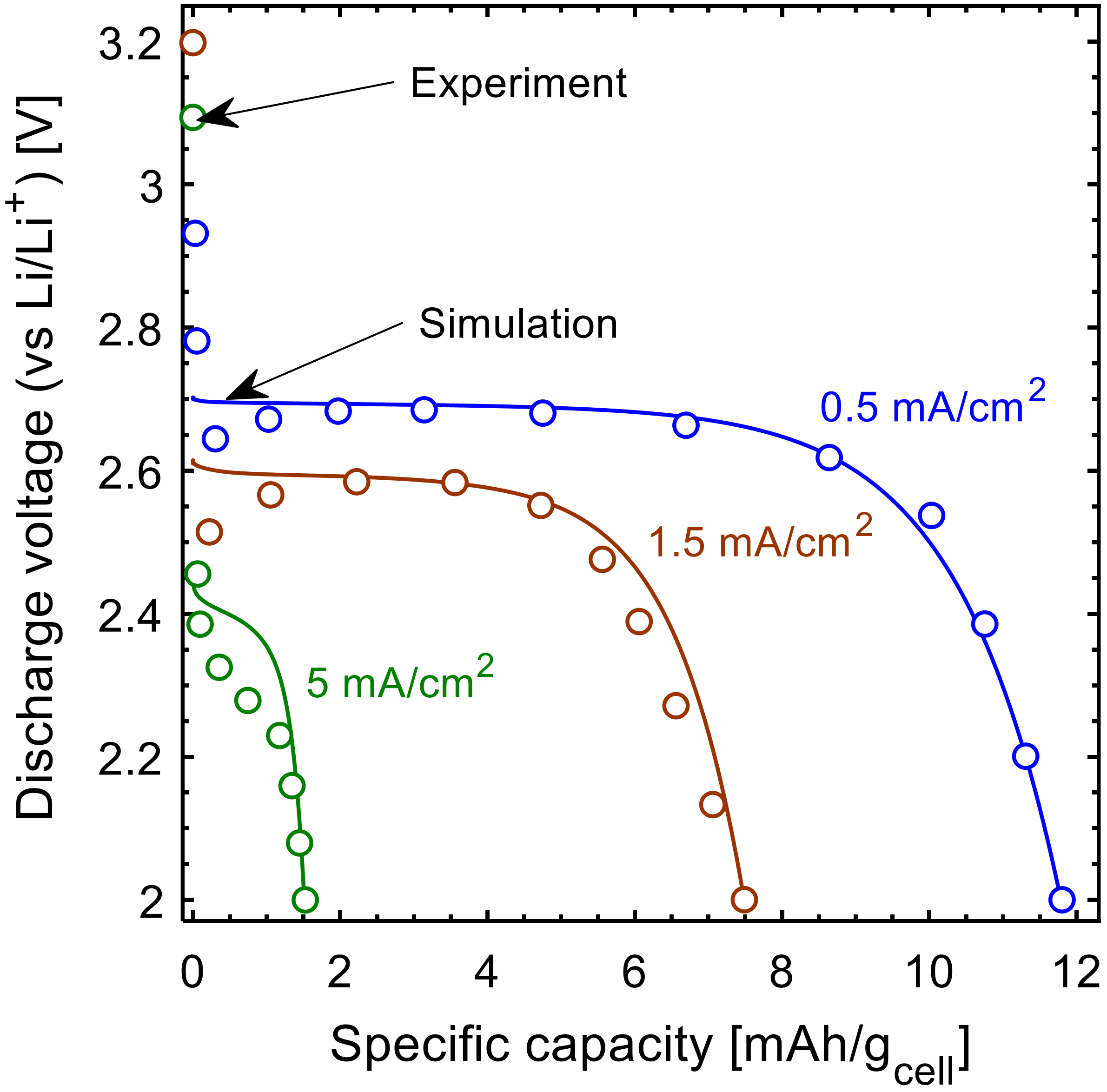
MD Simulations

Electrolyte

Experiments

Kinetics

Model Verification



Simulating cells for high power cell needs accurate electrolyte properties and current dependent kinetics

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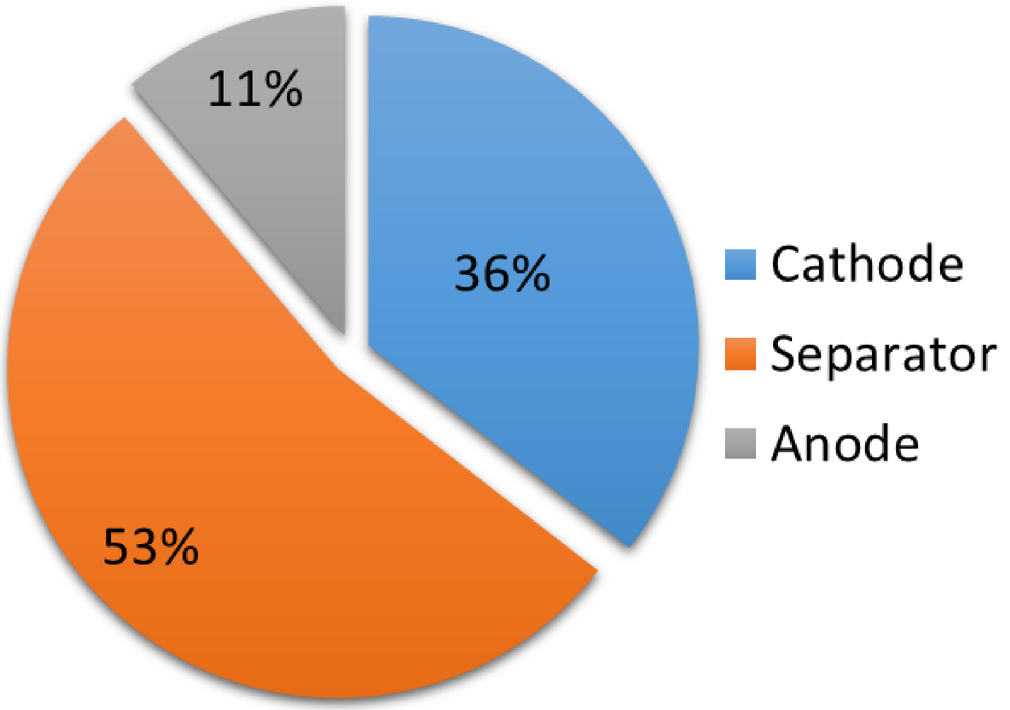
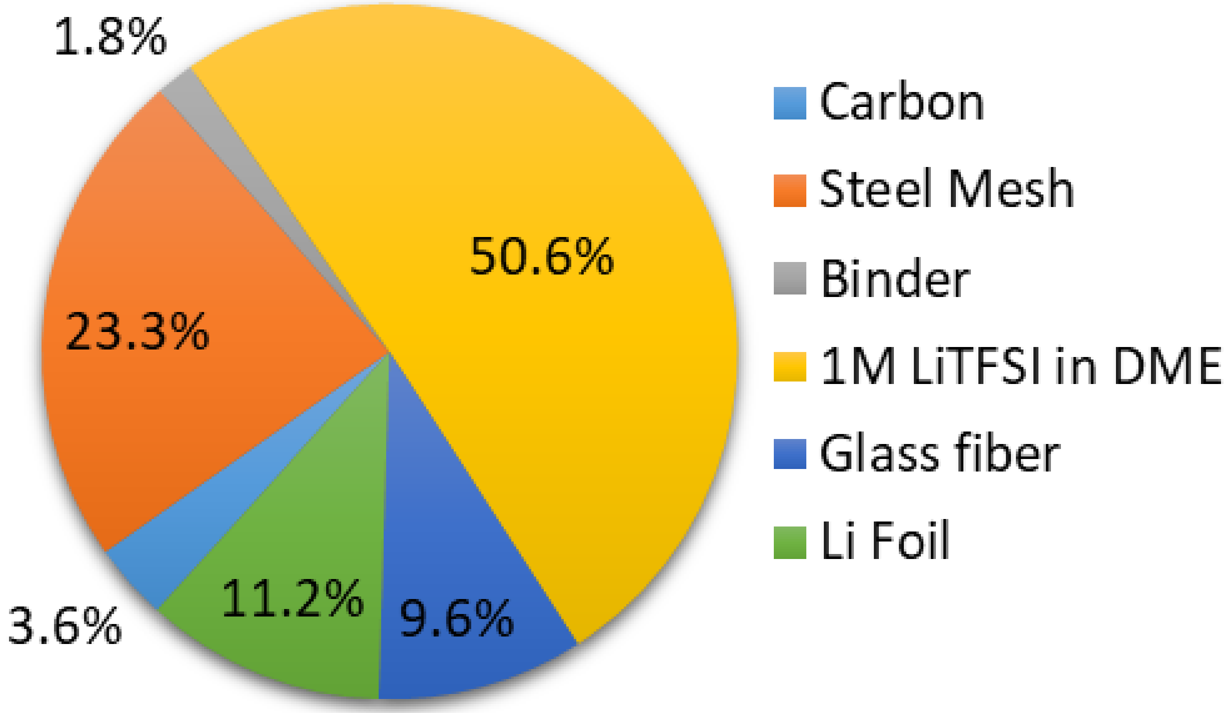
Electrochemical mass distribution



Cell mass distribution



Mass distribution separated into solid and liquid phases

All three components of Li-O_2 cell can be optimized to achieve high specific power

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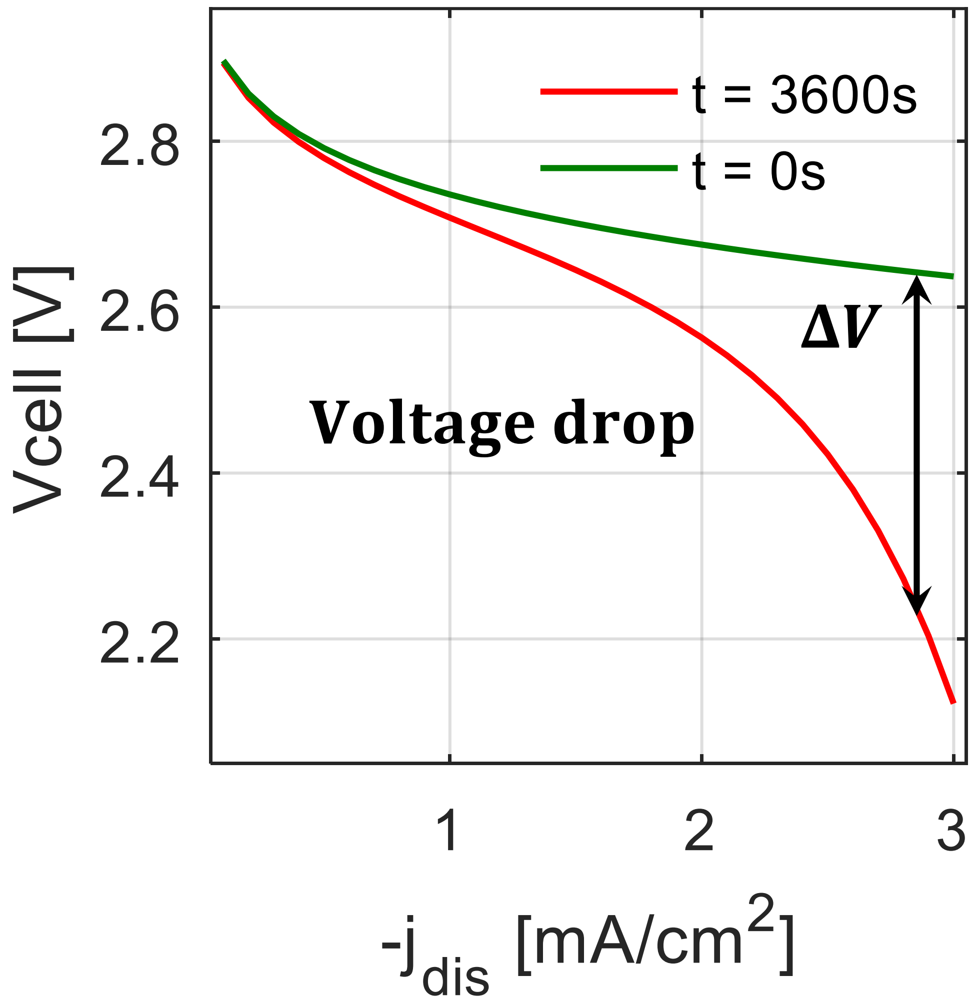
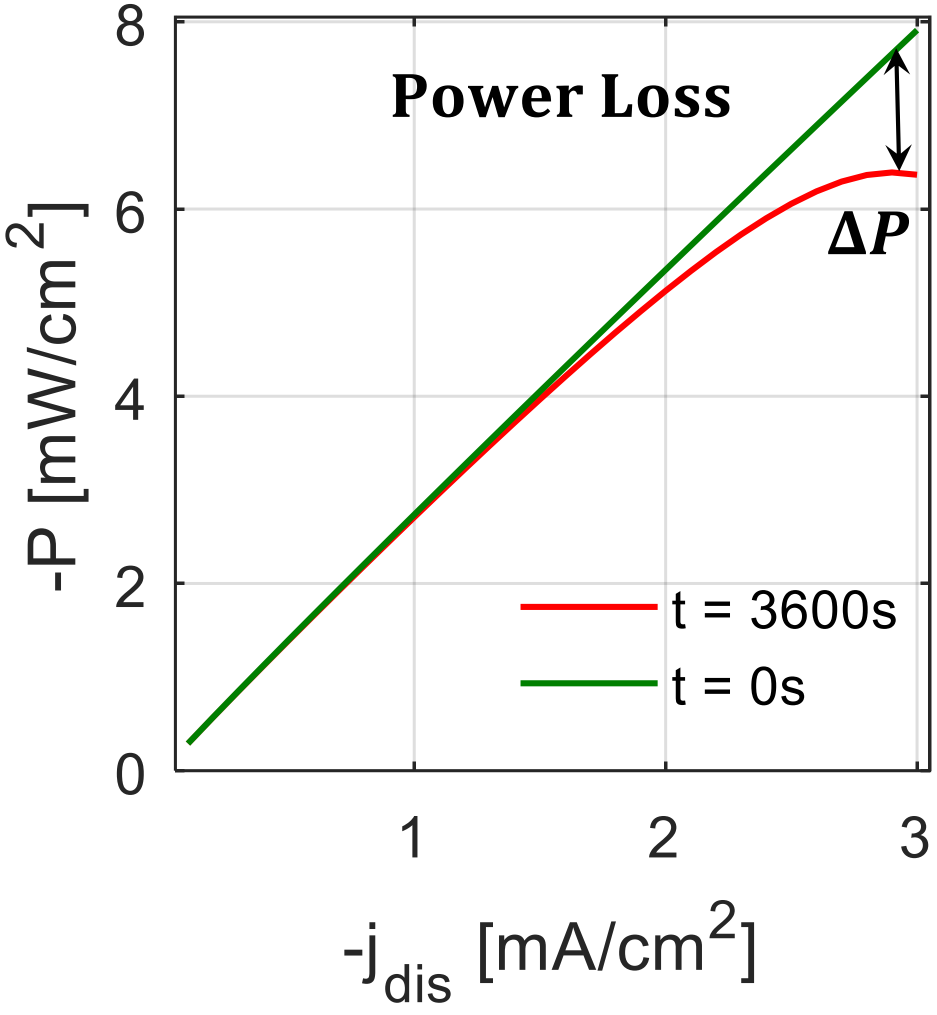
Polarization test: The effect on power

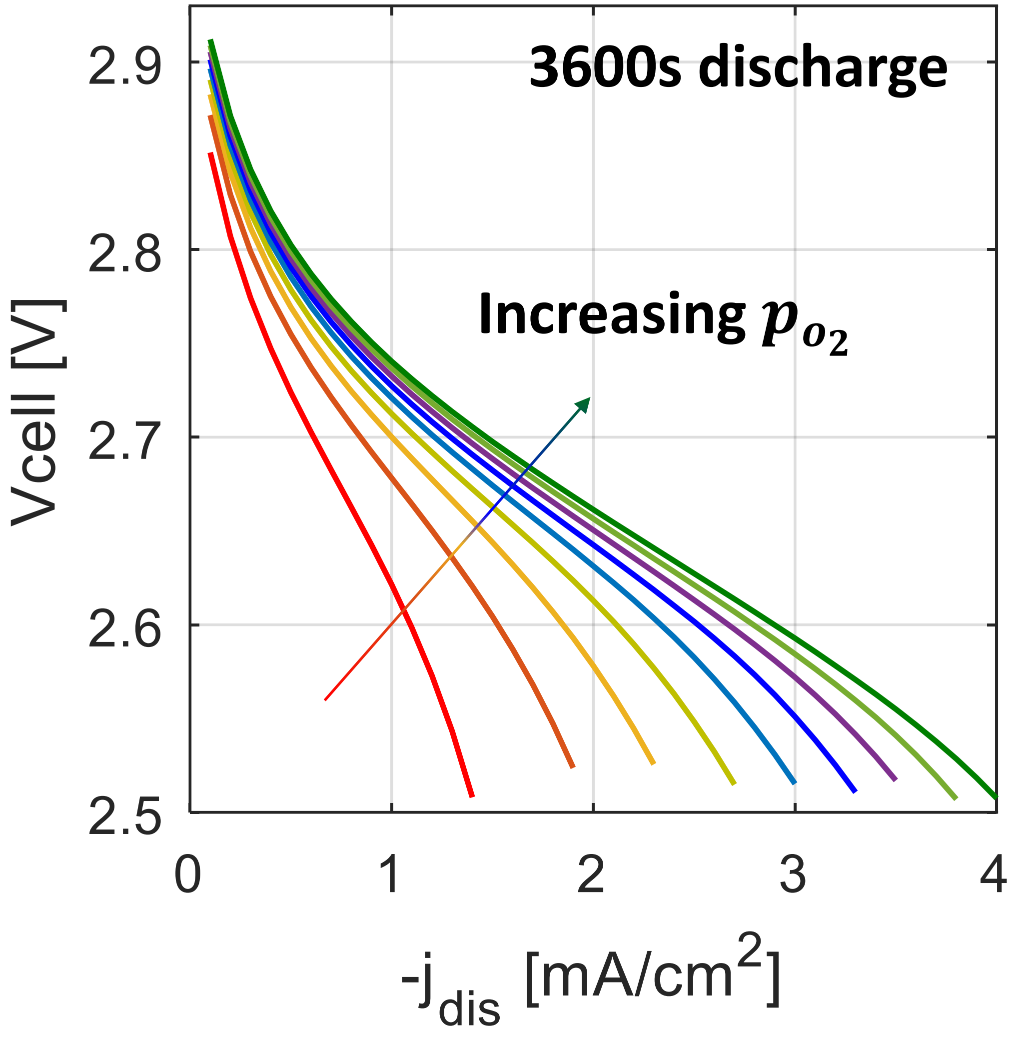


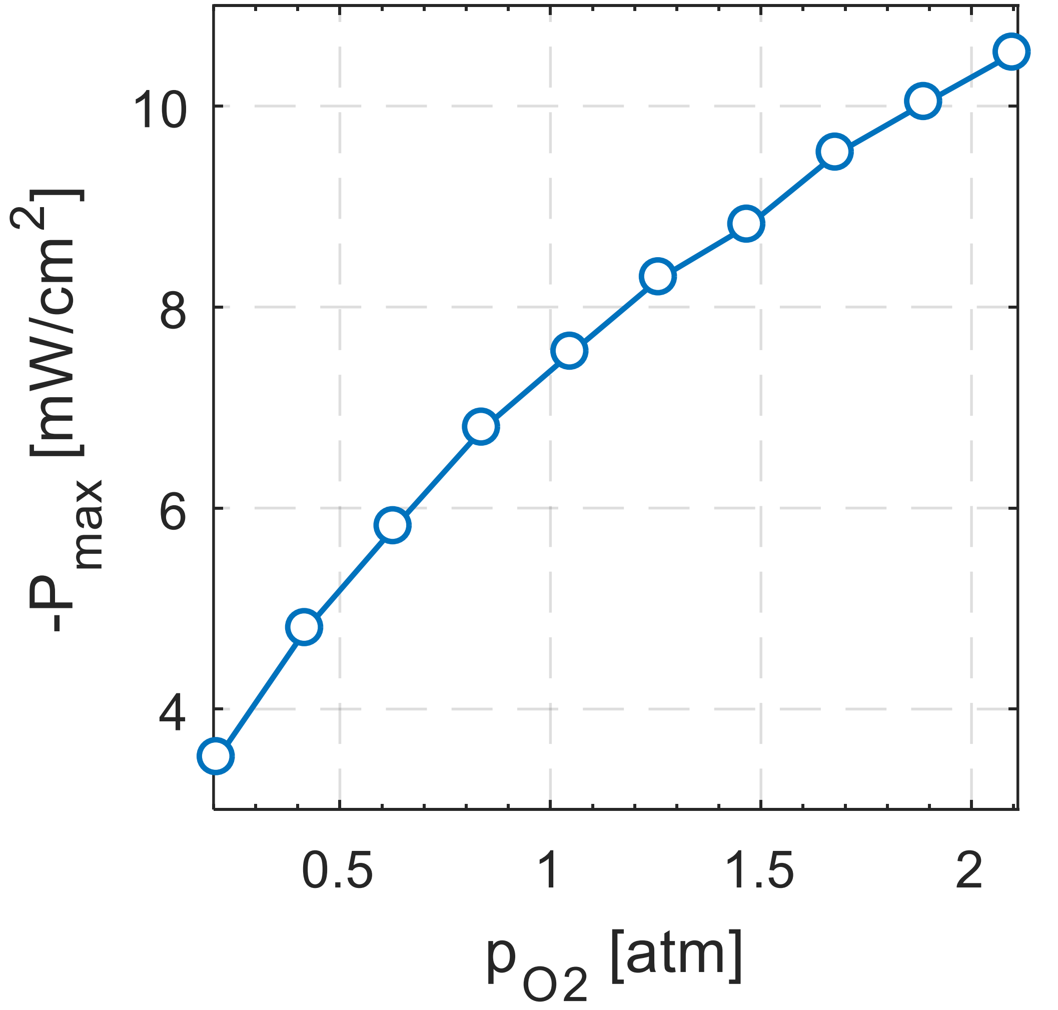


Operating at "high" current densities can lead to 25% power loss during 1hr discharge

Mehta *et. al.* in preparation, 2019

Polarization test: Oxygen Partial Pressure

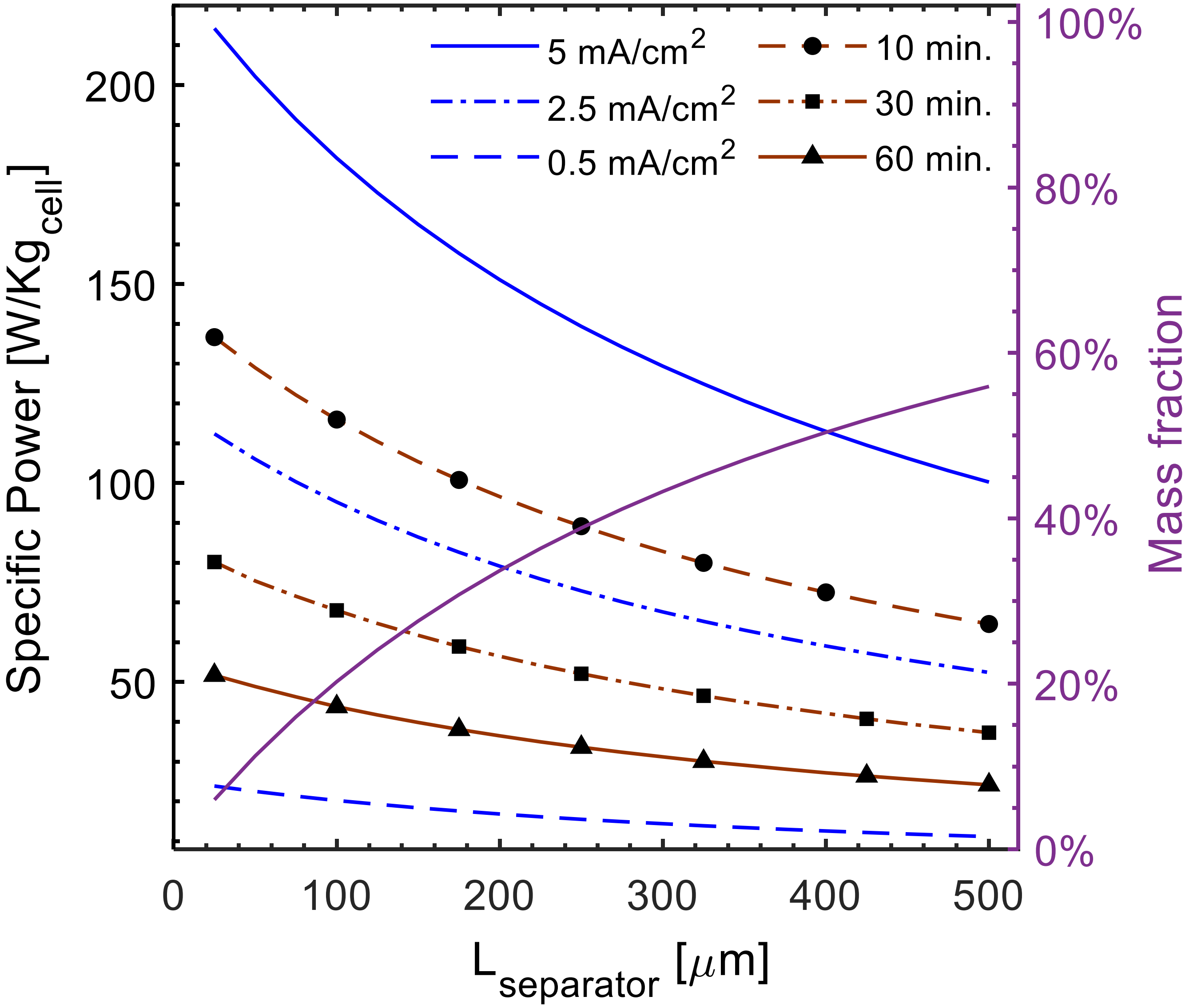




Increasing oxygen partial pressure improves power as well as non-electrochemical mass

Mehta *et. al.* in preparation, 2019

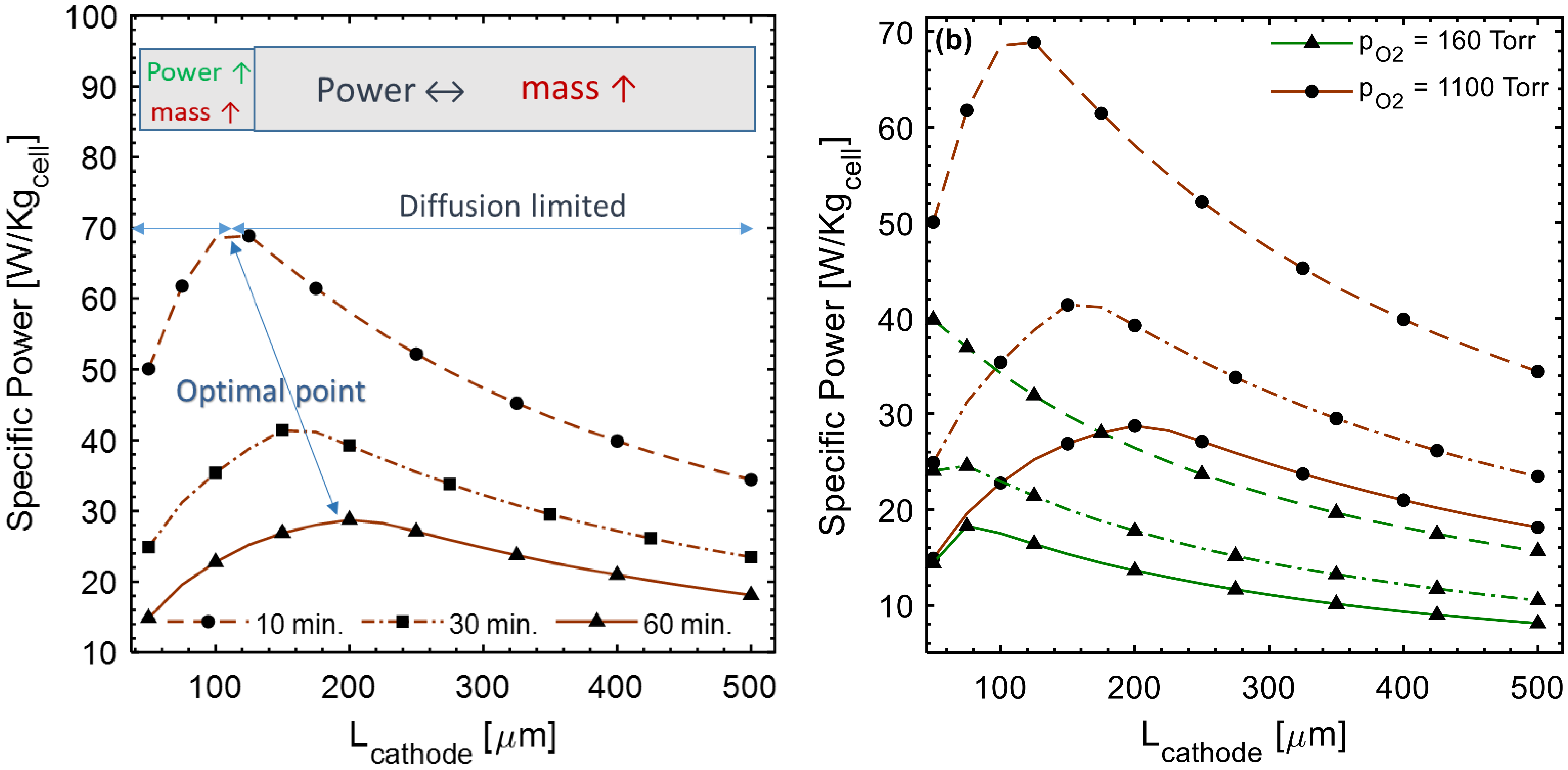
Influence of separator on performance



Separator does not contribute to battery performance at high current densities

Mehta *et. al.* in preparation, 2019

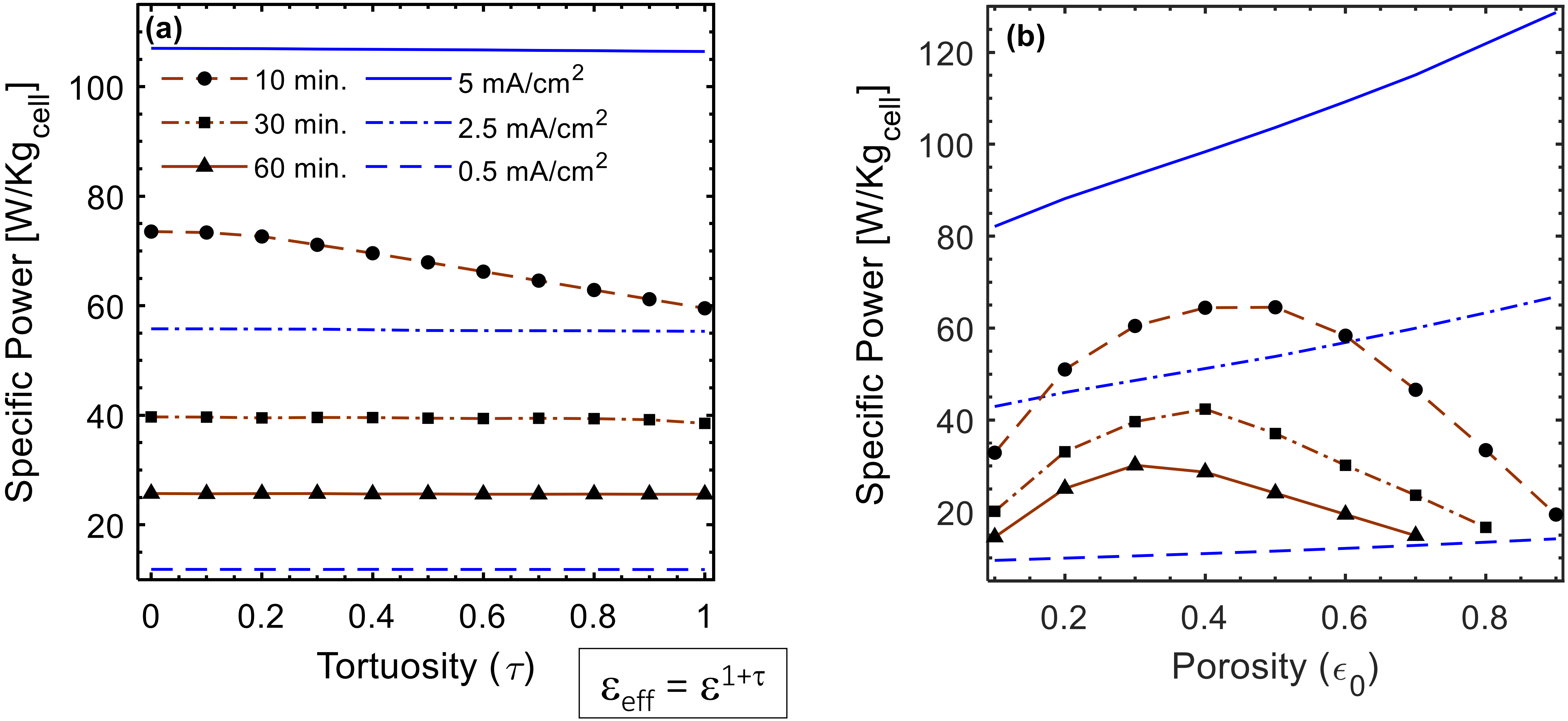
Effect of cathode thickness on performance



Optimal cathode thickness depends on operation conditions

Mehta *et. al.* in preparation, 2019

Influence of microstructure on performance

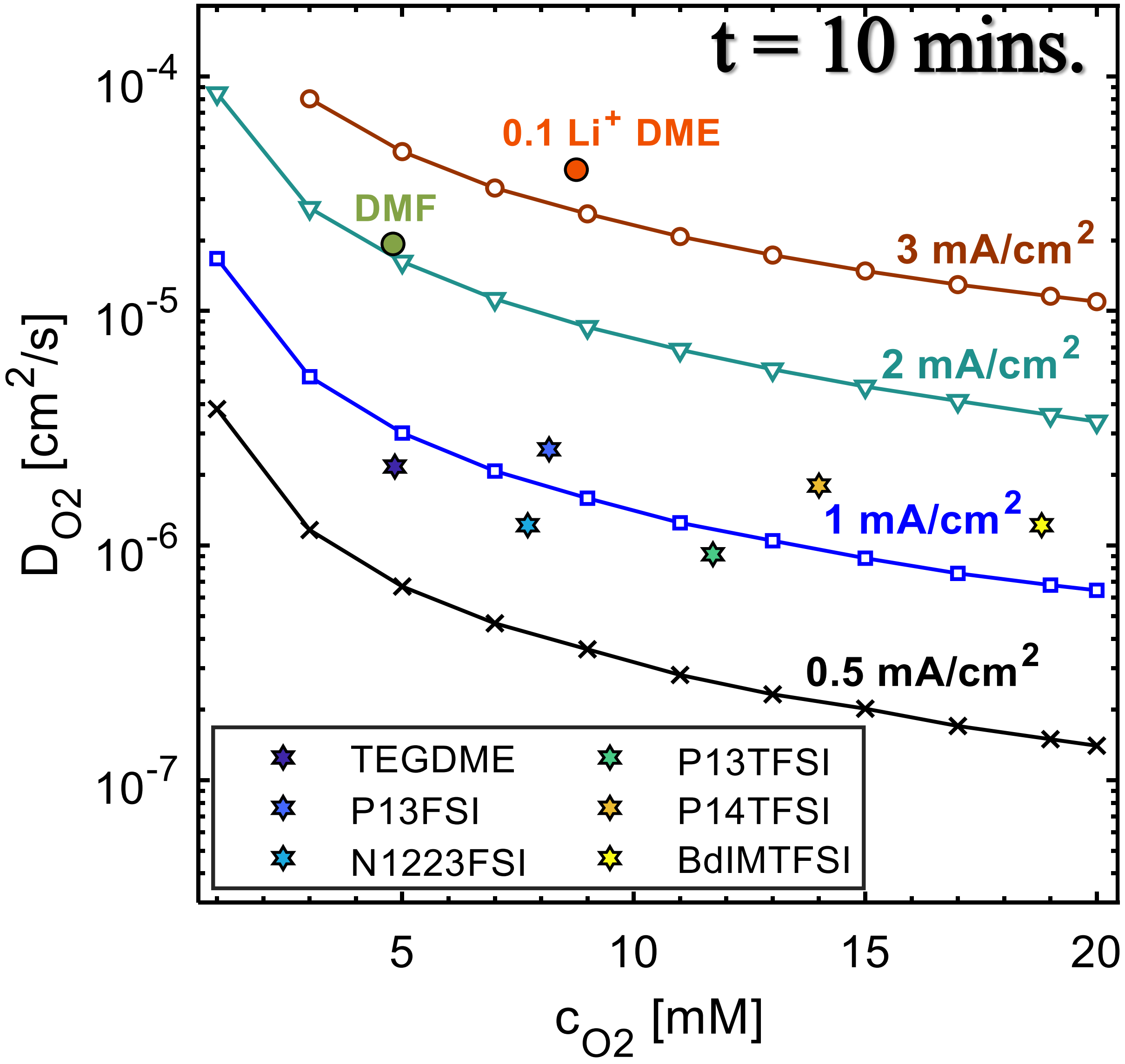


$$\varepsilon_{eff} = \varepsilon^{1+\tau}$$

Optimal values for porosity, particle size, and tortuosity depend on discharge current density and discharge time

Mehta *et. al.* in preparation, 2019

Influence of electrolyte properties



Diffusion requirements can be relaxed based by changing operating partial pressure and choosing lower salt concentration

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Simulation-based optimization (30 min.)

1 ● 0.3M LiTFSI P_{13}FSI

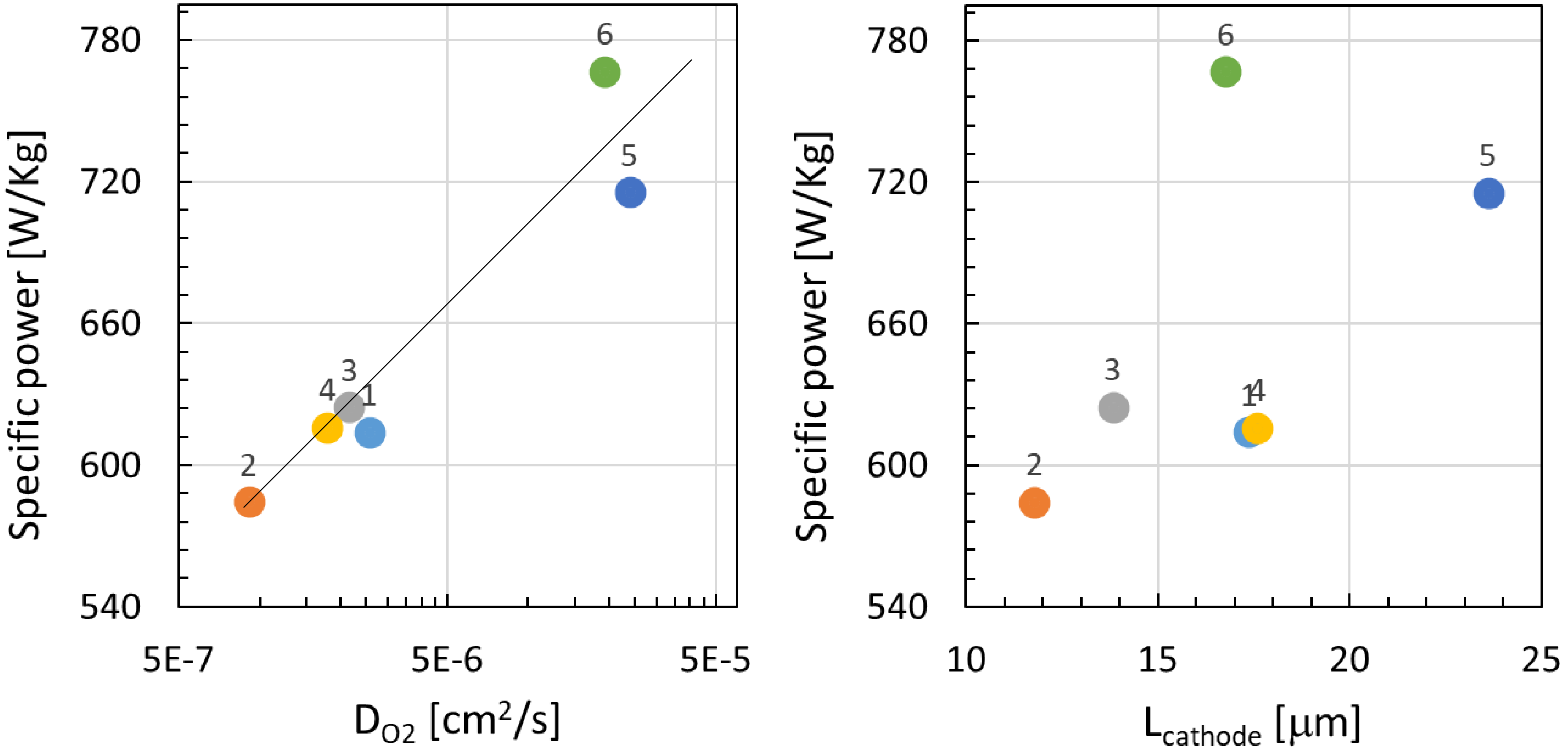
2 ● 0.3M LiTFSI P_{13}TFSI

3 ● 0.1M LiTFSI TEGDME

4 ● 0.3M LiTFSI P_{14}TFSI

5 ● 1M LiTFSI DME

6 ● 0.1 TEAP DMF



The oxygen diffusion length (under steady-state) determines cathode thickness and cathode mass

Thin and optimized cathodes, and better oxygen transport electrolytes can provide Li-O_2 for high-specific power cells

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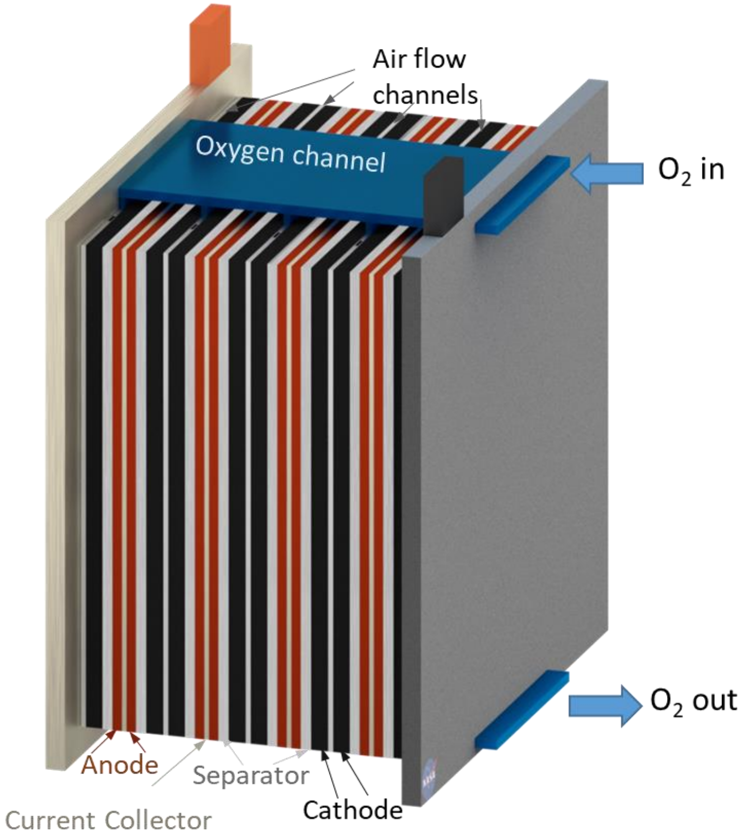
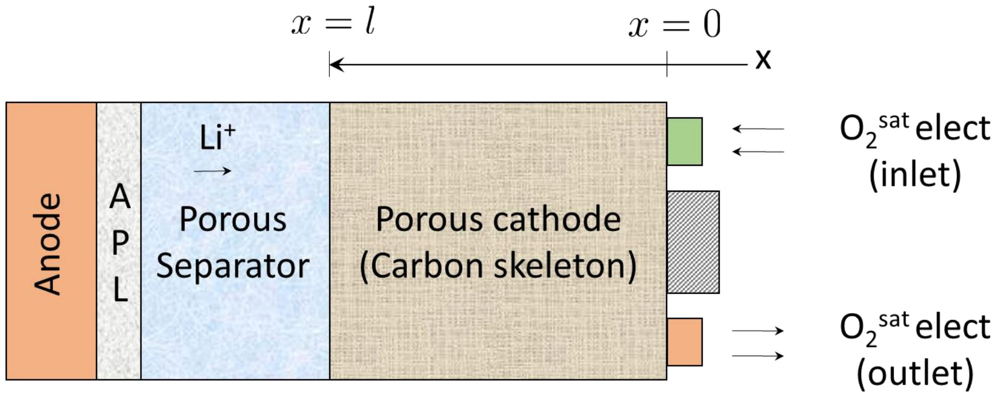
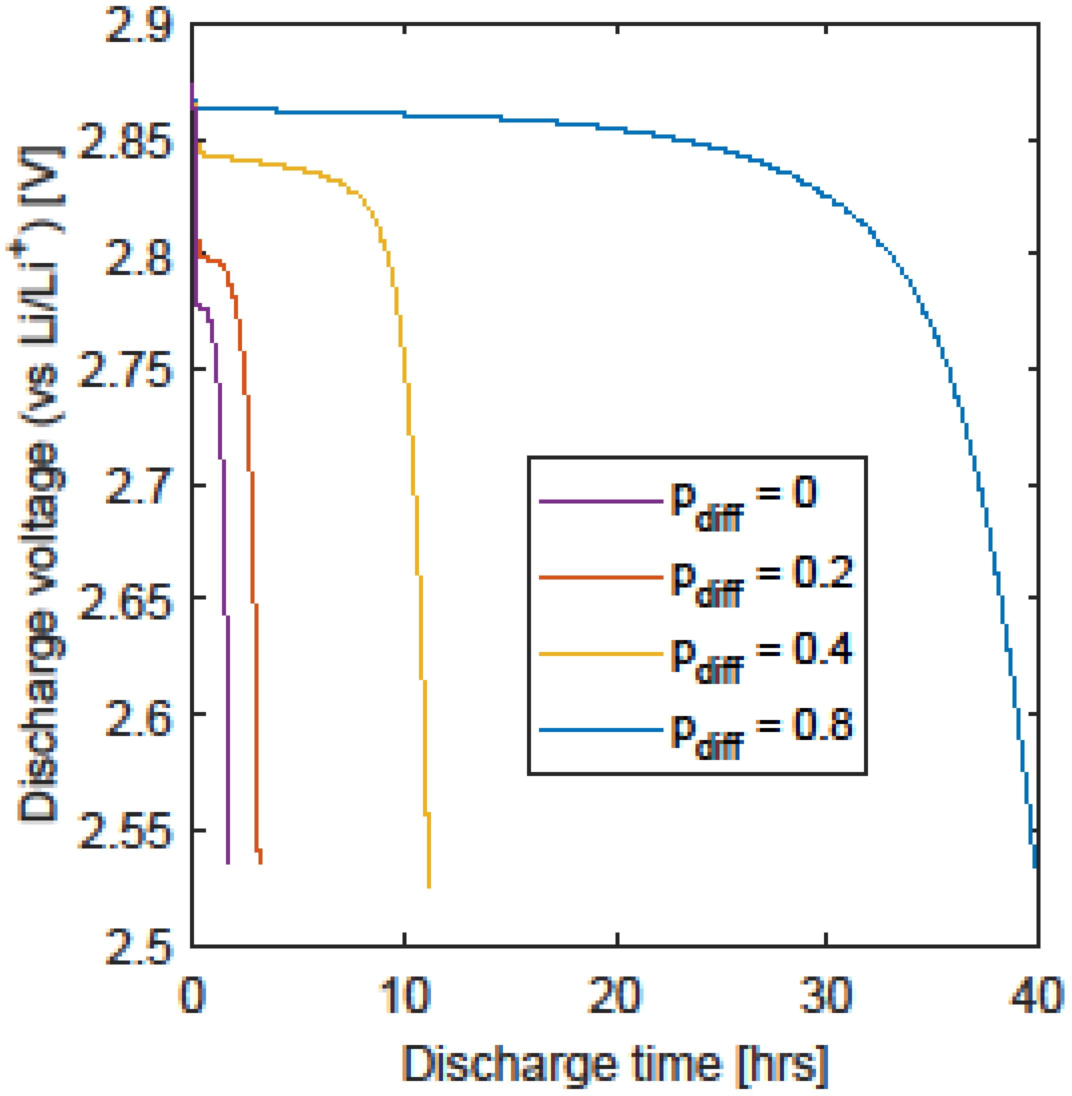
Pack level simulation (better optimization)







Active cathode design shows performance improvement but at a power cost of 5-30% for running external systems

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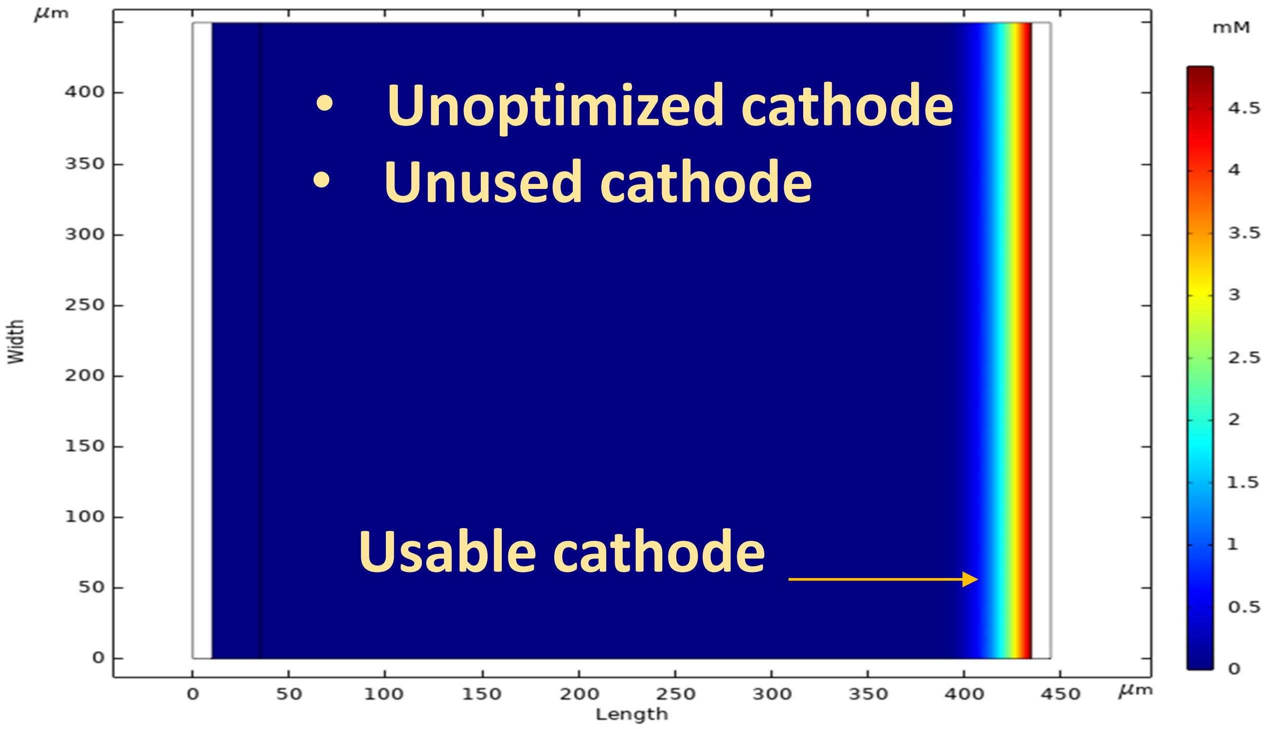
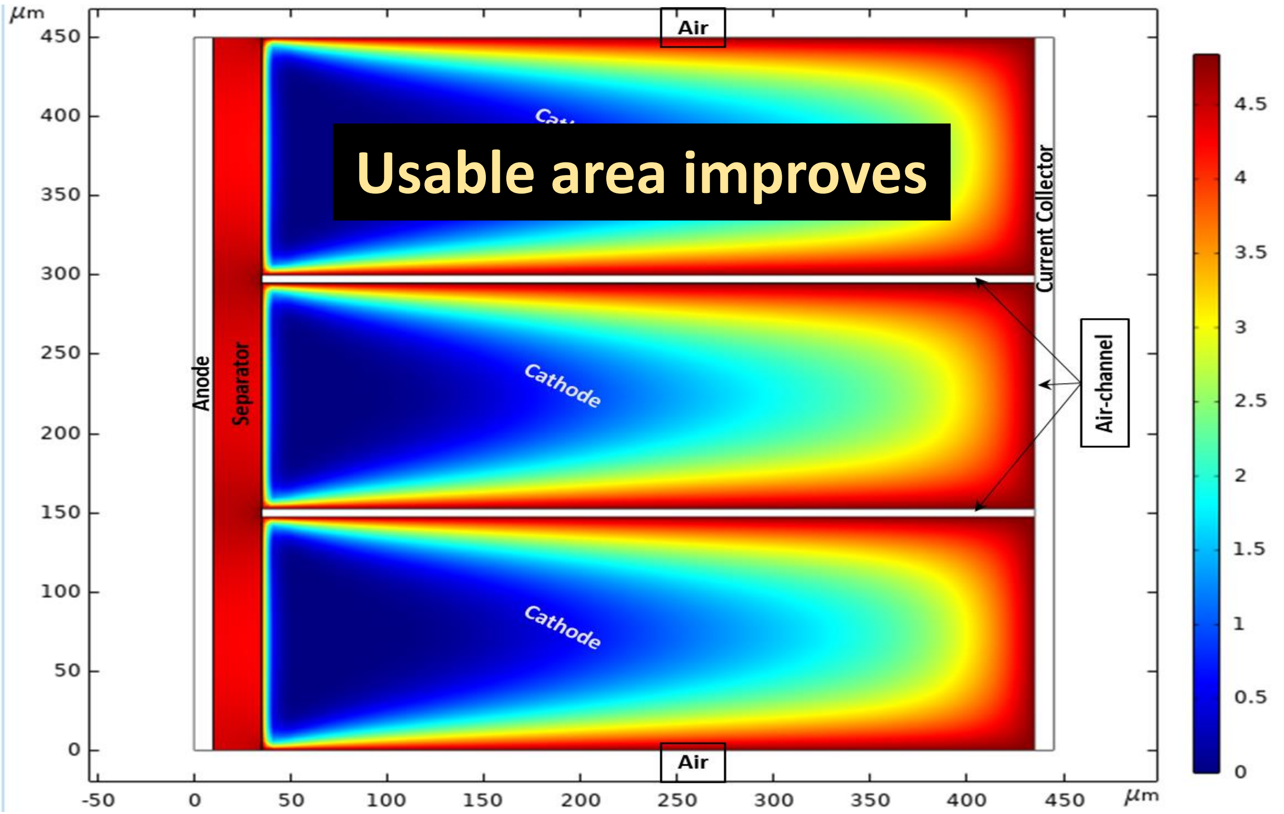
Pack level simulation (better optimization)

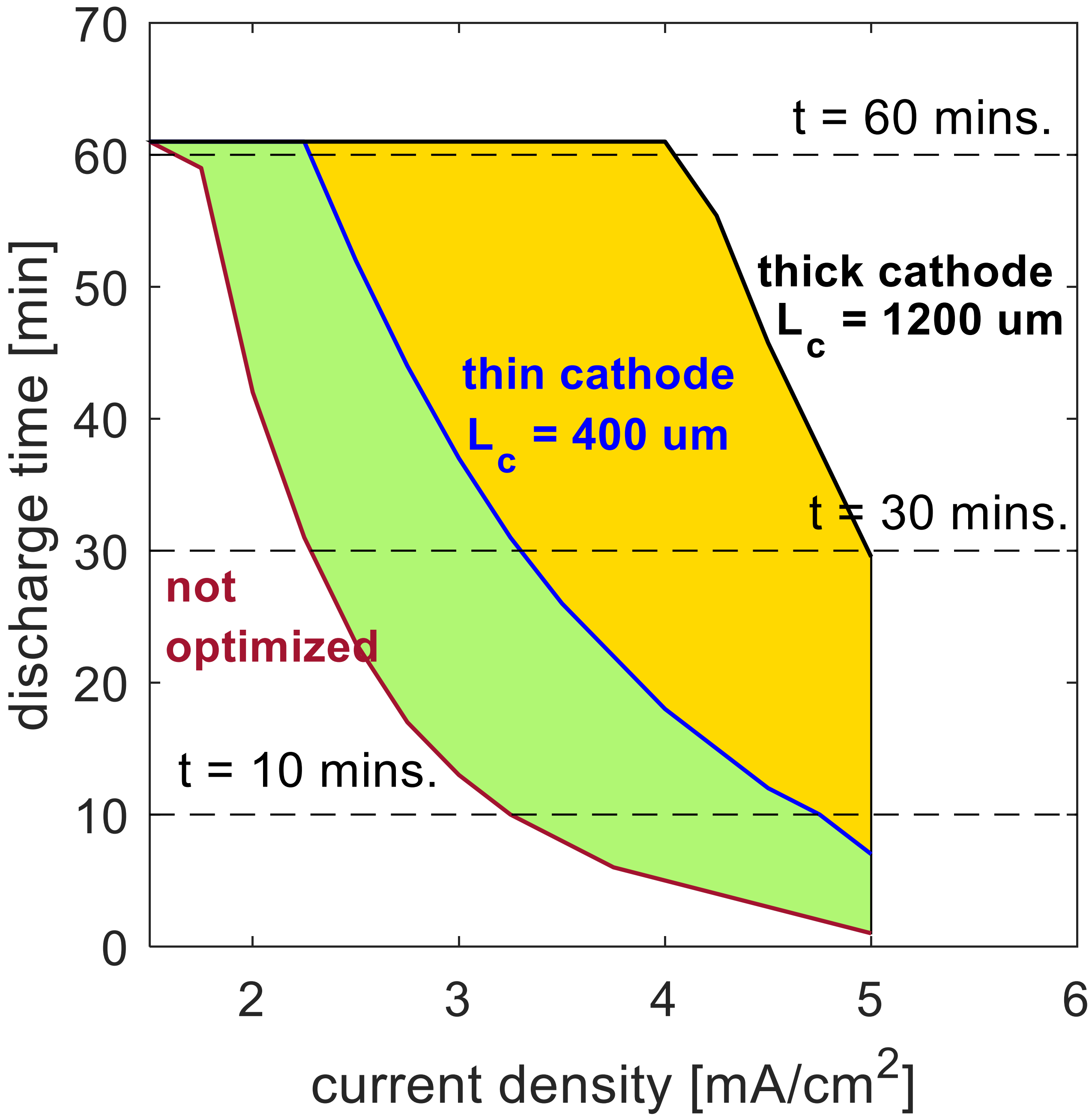


- Unoptimized cathode
- Unused cathode

Usable cathode



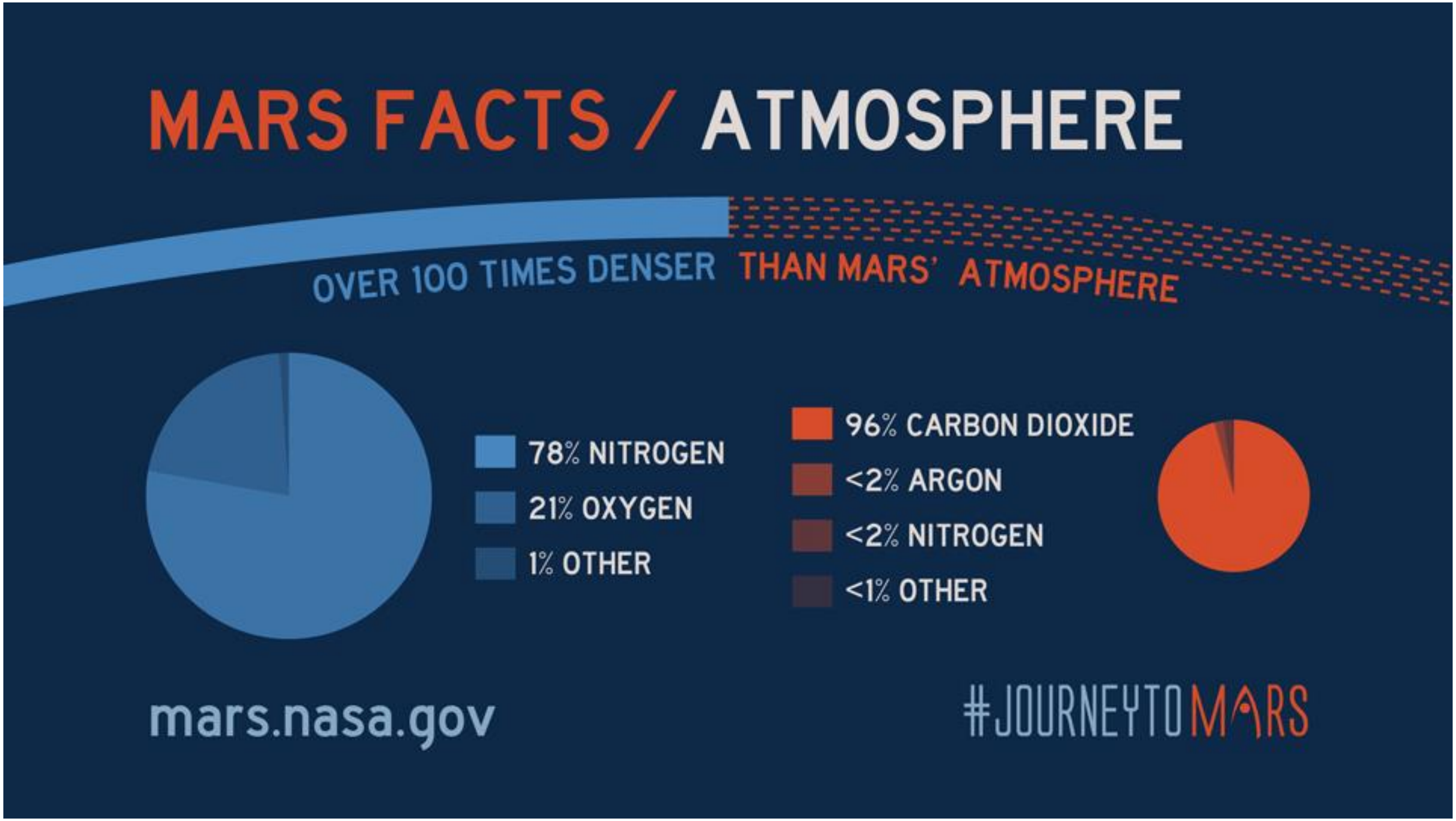
Usable area improves



Better cathode utilization improves discharge time at high current 30x

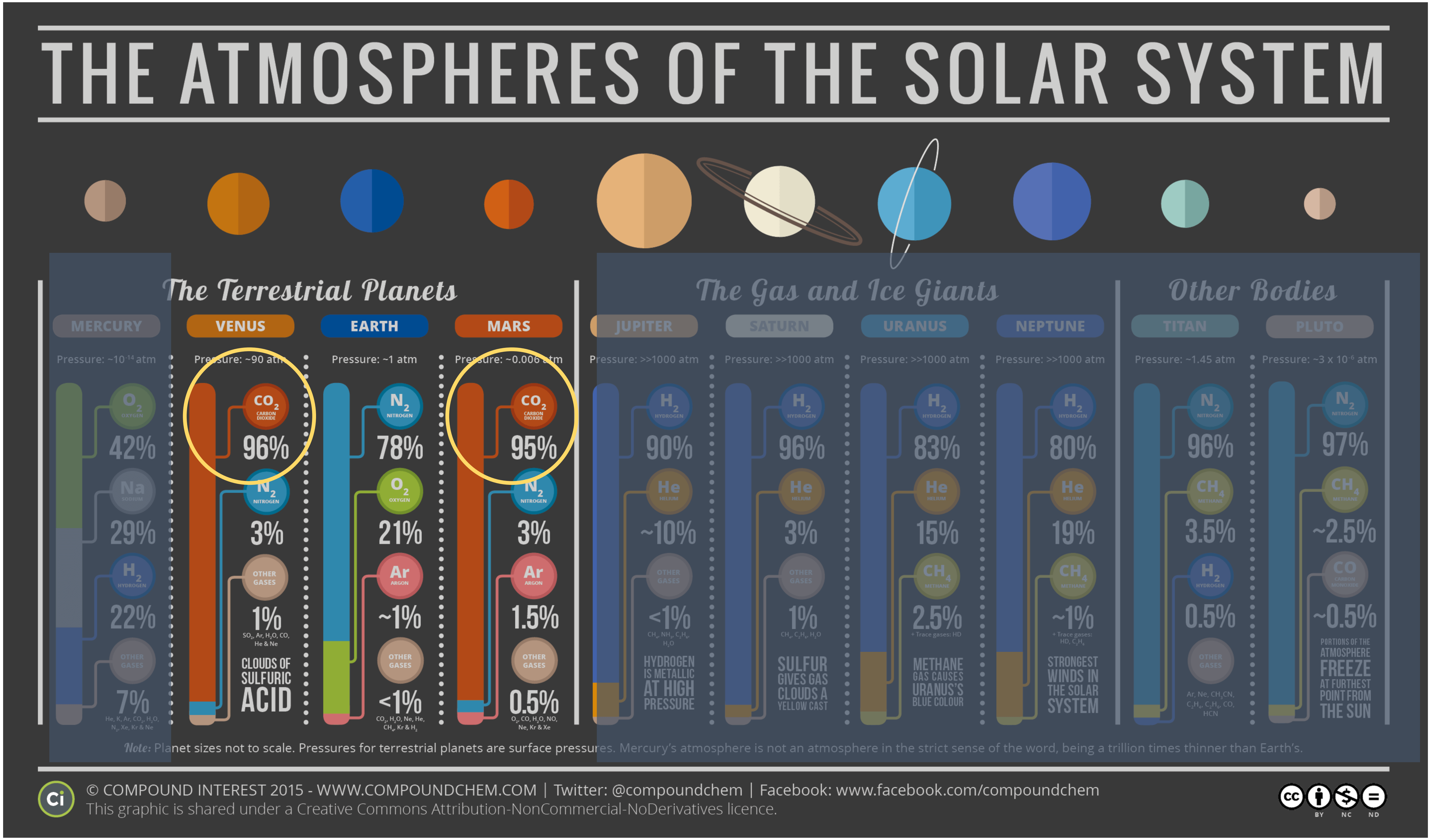
Mehta *et. al.* in preparation, 2019

Batteries for Space (Motivation)



Courtesy NASA/JPL-Caltech

Batteries for Space (Motivation)



THE ATMOSPHERES OF THE SOLAR SYSTEM

Note: Planet sizes not to scale. Pressures for terrestrial planets are surface pressures. Mercury's atmosphere is not an atmosphere in the strict sense of the word, being a trillion times thinner than Earth's.

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Requirements for “Space” Batteries

Operating Temperature: -170ºC – 200ºC

Specific Energy (Reversible): > 500 Wh/Kg

Extremely Low Self-discharge (0-volt)

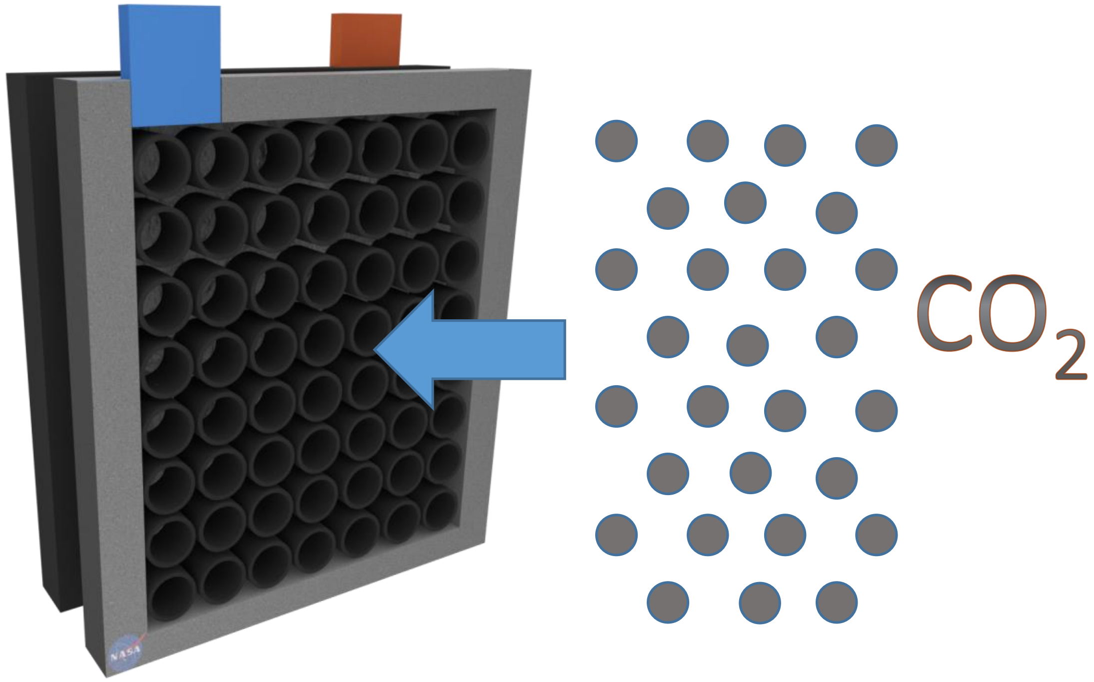
Battery System needs to be low mass and volume

Current systems need temperature regulation for optimal performance

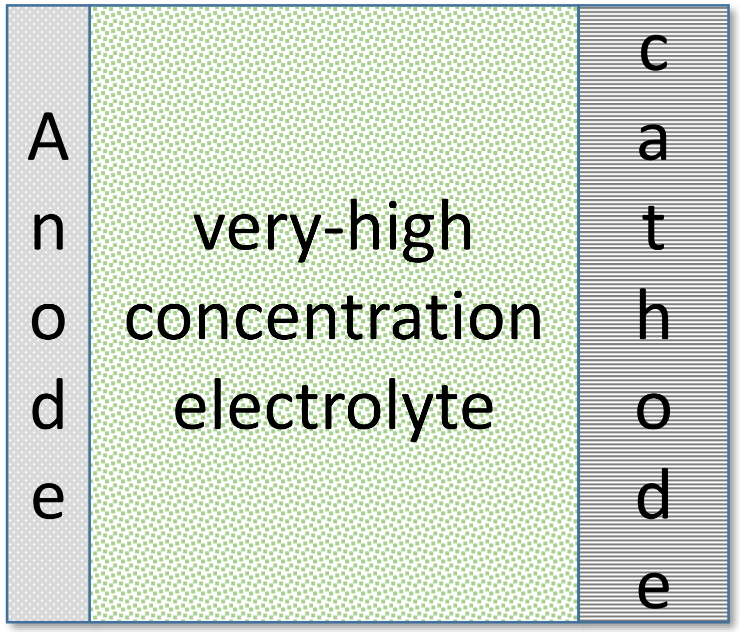
Batteries need to be Safe and Reliable

Emerging Battery Technologies for Space

Li-CO_2 Battery:



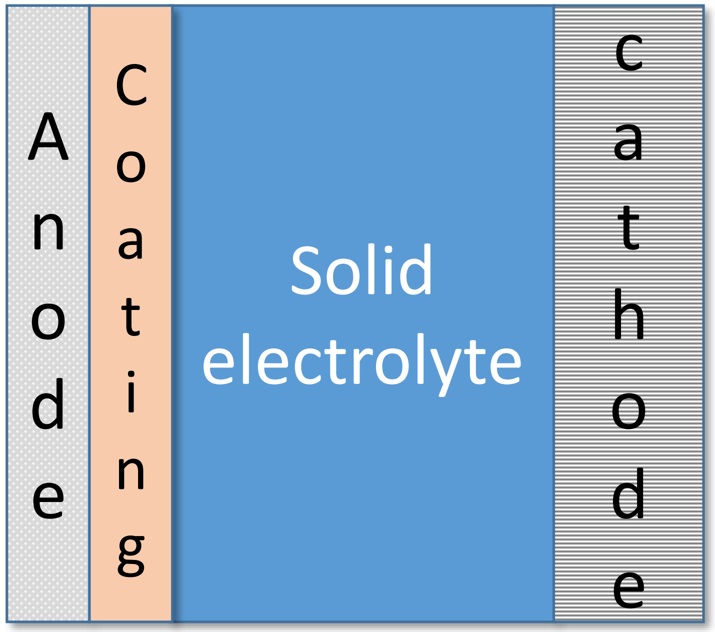
Solvent-in-Salt Battery


Anode

very-high concentration electrolyte

cathode

Solid-State Battery:


Anode

Coating

Solid electrolyte

cathode

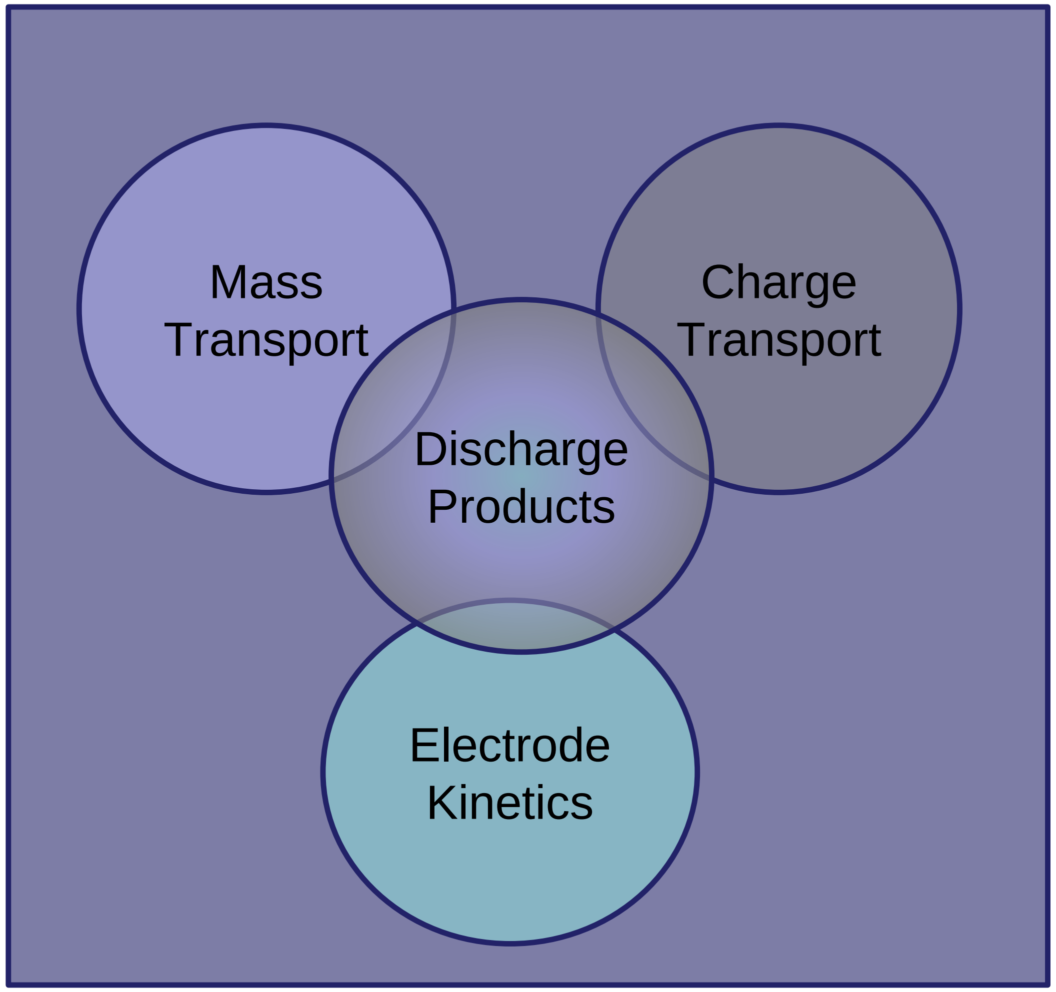
Utilizing Venus and Mars atmosphere

Li-CO_2 Battery:

$$4\,\mathrm{Li}^{+} + 3\,\mathrm{CO}_2 + 4\,\mathrm{e}^{-} \longrightarrow 2\,\mathrm{Li}_2\mathrm{CO}_3 + \mathrm{C}\,(E^{\circ} = 2.8\,\mathrm{V})$$

$$4\,\mathrm{Li}^{+} + \mathrm{CO}_2 + 4\,\mathrm{e}^{-} \longrightarrow 2\,\mathrm{Li}_2\mathrm{O} + \mathrm{C}\,(E^{\circ} = 1.89\,\mathrm{V})$$

Modeling Framework identical to Li-O_2

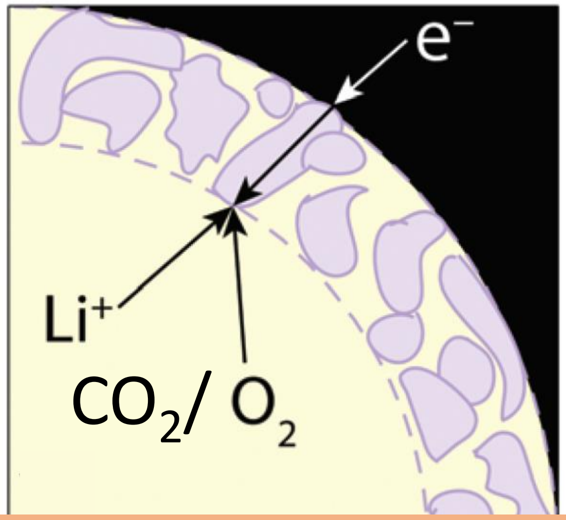


- OCV similar to Li-O_2 chemistry
- The reaction pathway changes below 1.89V
- Lithium Carbonate is more insulating than Li_2O_2
- The kinetics are facile than Li-O_2

Modeling similarities with Li-O_2 chemistry

Morphology of Discharge product

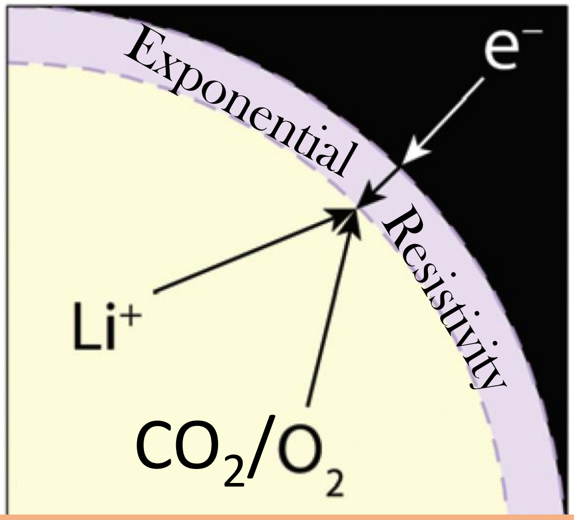
Particle deposition



Polymer Electrolytes (CO_2)

Low current density (O_2)

Film deposition



Liquid Electrolytes (CO_2)

High current density (O_2)

Liu, J., Rahimian, S. K. & Monroe, C. W. *Physical Chemistry Chemical Physics* **18**, 22840–22851 (2016).

Electrolyte Transport

Different Solubility: 125mM (CO_2) > 2mM (O_2)

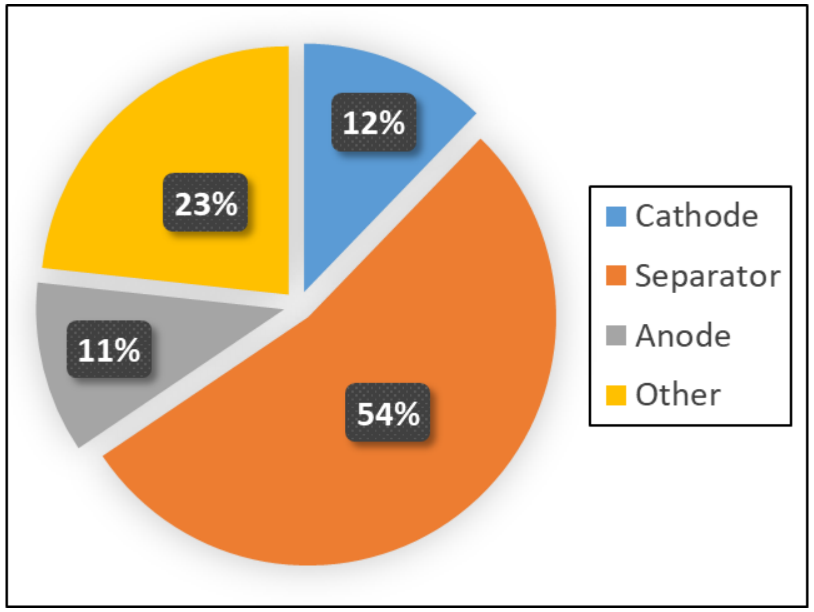
Similar Diffusivity: 10^{-5} cm^2/s (CO_2) ≈ 2×10^{-5} cm^2/s

Wadhawan, J. D., Welford, P. J., McPeak, H. B., Hahn, C. E. W. & Compton, R. G. *Sensors and Actuators B: Chemical* **88**, 40–52 (2003).

Summary

1. Batteries for Aviation

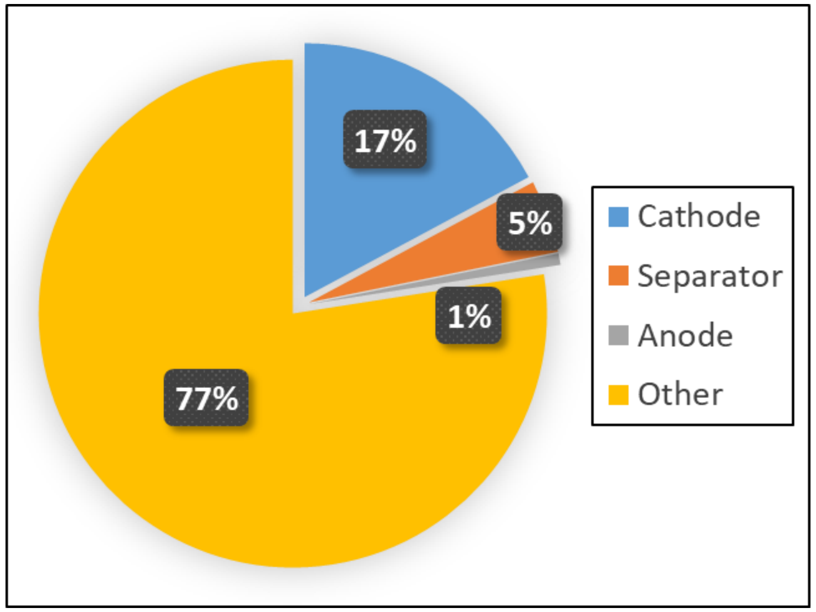
- Physics-based models can guide cell and pack designs for aviation batteries
- Both current density and cell mass needs to be optimized for high specific power
- Optimal cell design changes based on discharge time, discharge current density, and operating conditions



Laboratory cell (not optimized)

2. Batteries for Space

- Physics-based models for emerging chemistries need to be developed
- Models on Li-O_2 can be ported to simulate Li-CO_2 batteries for Mars and Venus



Cell optimized for low electrochemical mass

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