Impact of High-Performance Computational Analysis on Design of Energy Storage Devices

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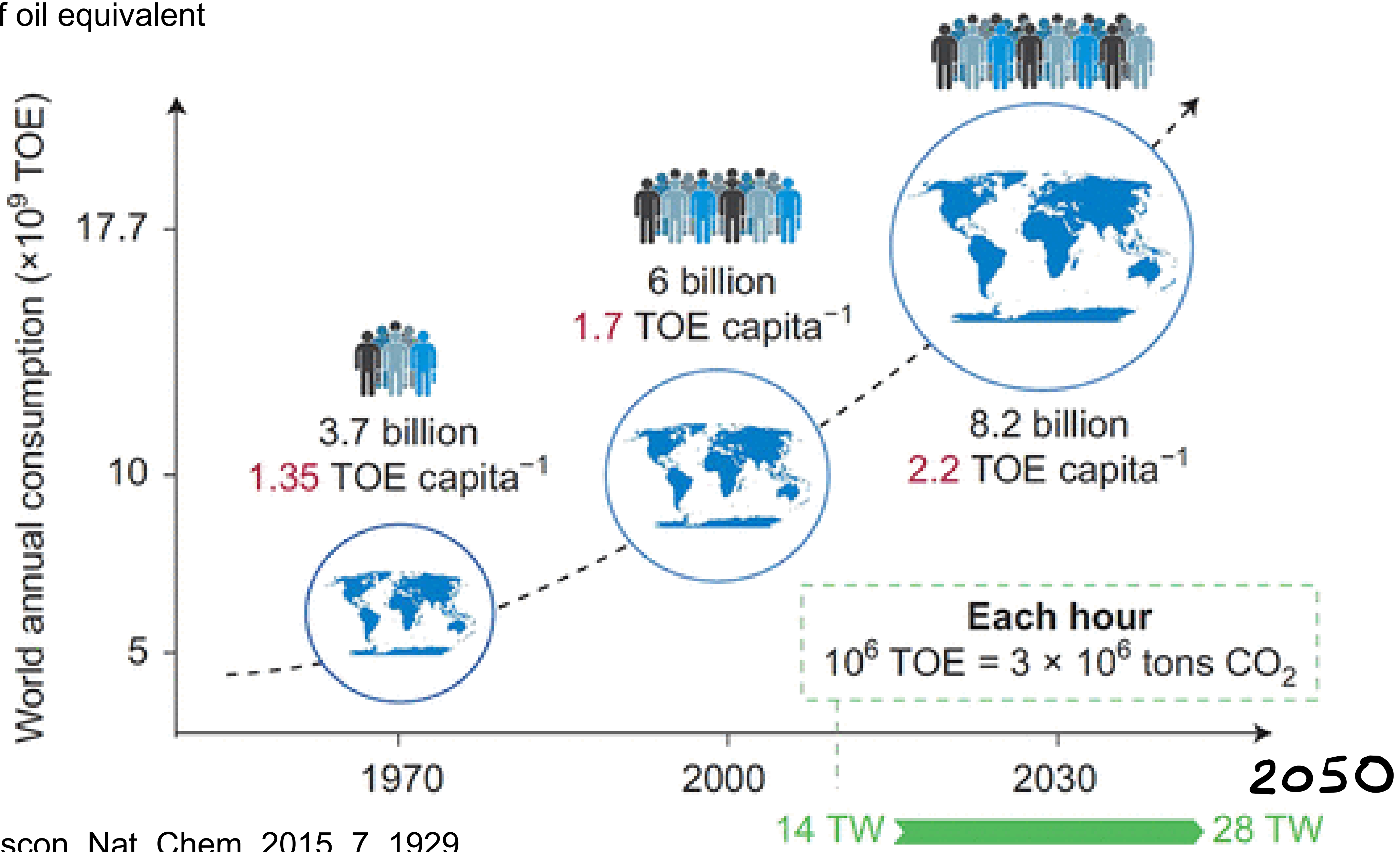
Forecast of Energy Consumption up to ~ 2050

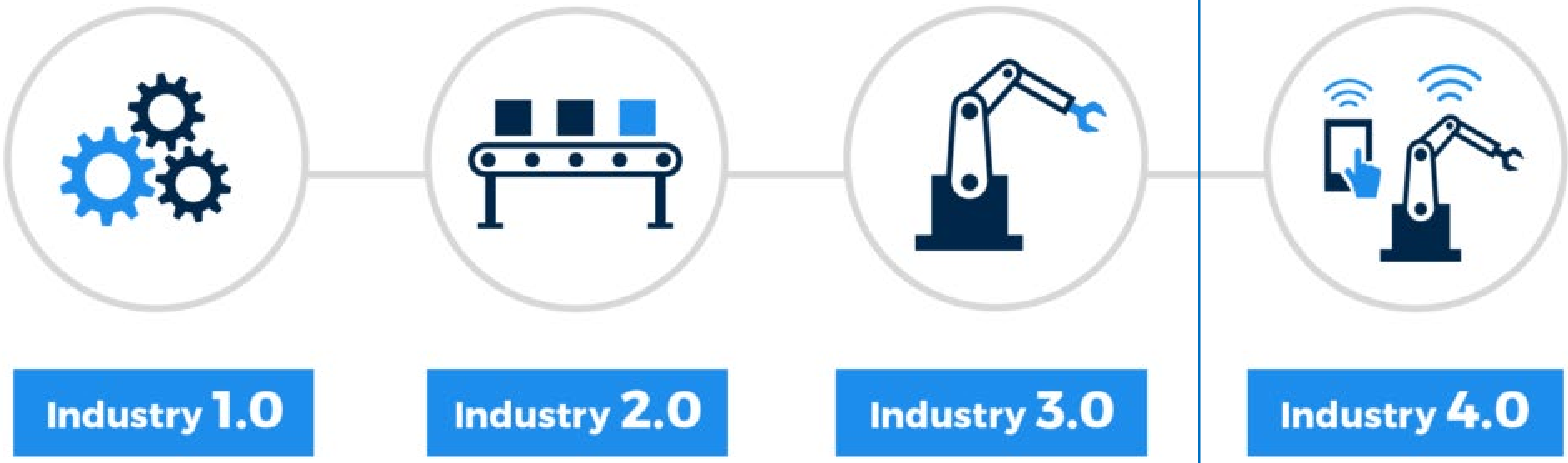
TOE = ton of oil equivalent



Larcher & Tarascon, Nat. Chem. 2015, 7, 1929

Technology Evolution and Energy Storage Needs

- steam engine (1st) industrialization
- electricity and mass production (2nd)
- computers (3rd) processing and sharing information
- automation (4th)



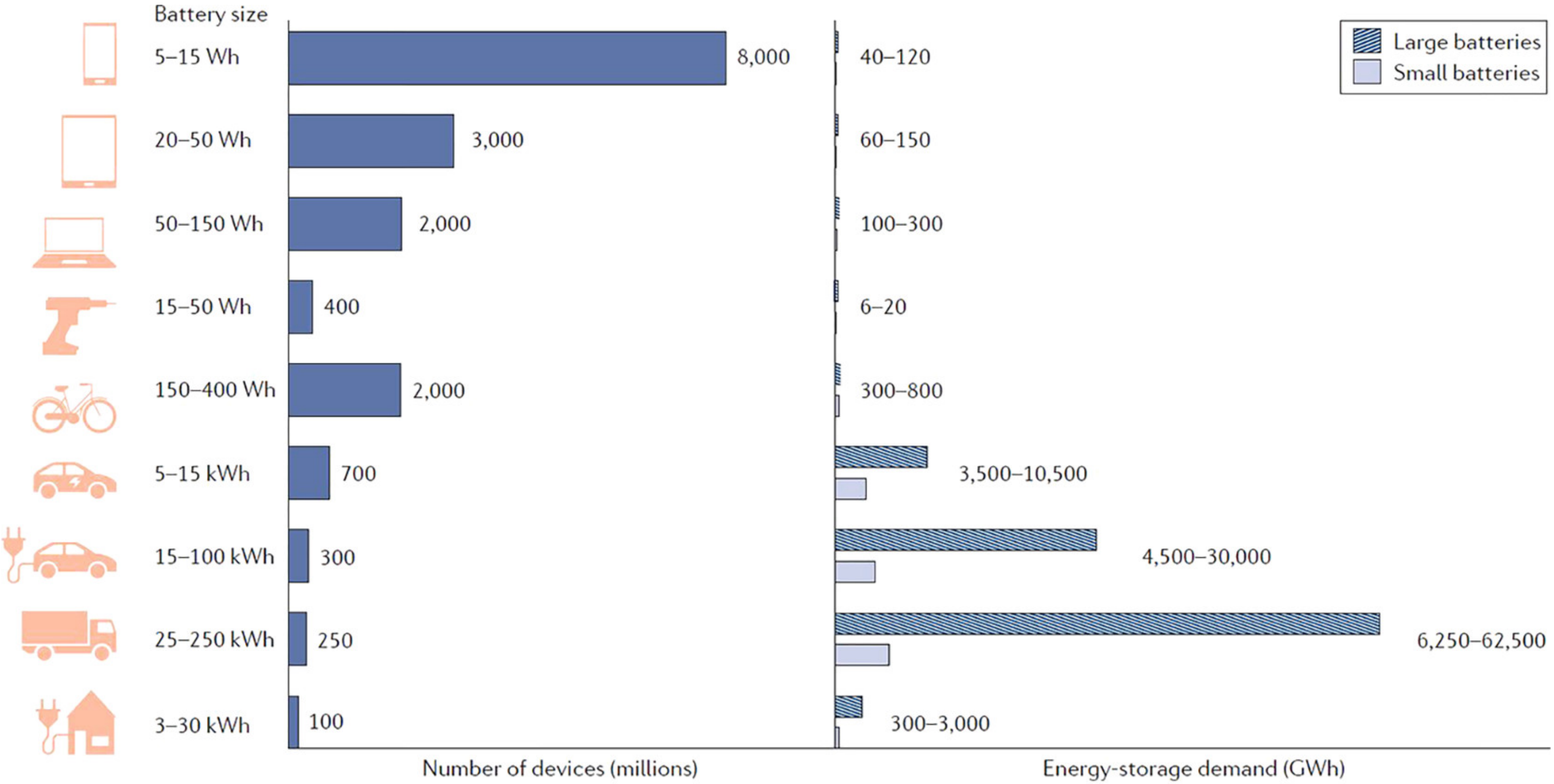
innovations
for autonomous
vehicles,
Internet of Things,
robotics,
drones…

Renewable Energies Need Batteries

Vistra Energy
(California)
has the largest
battery storage
system since Jan 2021
300MW/1,200 MWh

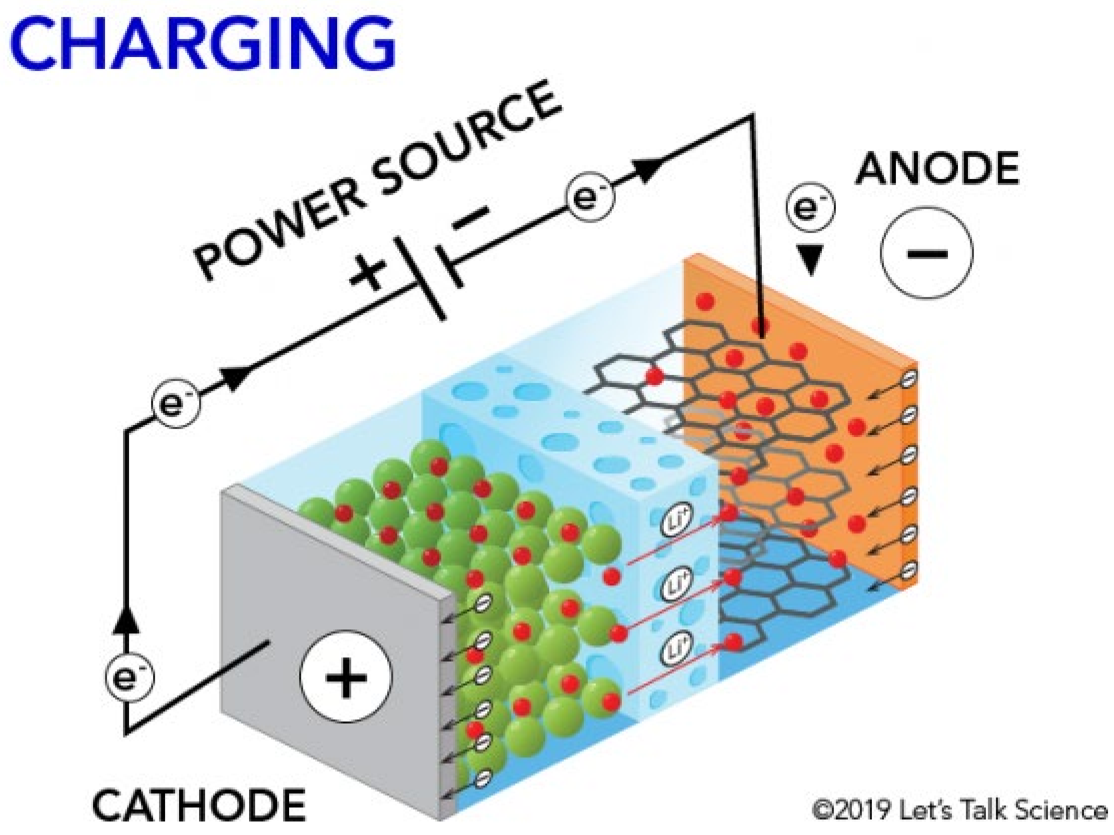
Southern Australia's Hornsdale Power Reserve 150MW/194MWh
(https://www.bbc.com/future/article/20201217-renewable-power-the-worlds-largest-battery)

Energy storage: # of devices; battery size



Larcher & Tarascon, Nat. Chem. 2015, 7, 1929

Energy stored during charge and released to an external load during discharge in a typical Li-ion battery



electrode materials

electrolyte materials

ion transport

electron transport

out of equilibrium process!!!

Current Status of Commercial Batteries: Materials Issues

- Shortage of metals (Li, Co, Ni, and others)
- Available low cost, abundant, non-toxic materials (Na, Mg, Ca, Al) → significant new challenges
- Electrolytes: cost, effectiveness (design)

Other Significant Challenges

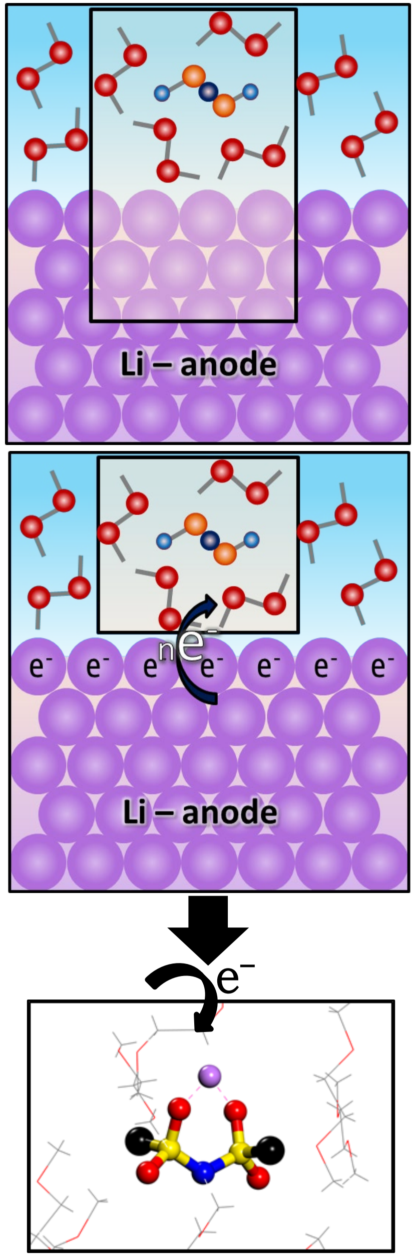
- Interfacial behavior
- Degradation, cycling, lifetimes
- Fast charge
- Cost
- Safety
- Mechanical stability
- Recycling materials
- Operation under wide range of temperatures
- Lack of understanding of interactions among battery components (anode, cathode, electrolyte)
- Effects of micro or nanostructure in anodes, cathodes, interfaces, interphases

Intrinsically out of equilibrium system requires "holistic" approaches, instead of "patching solutions"

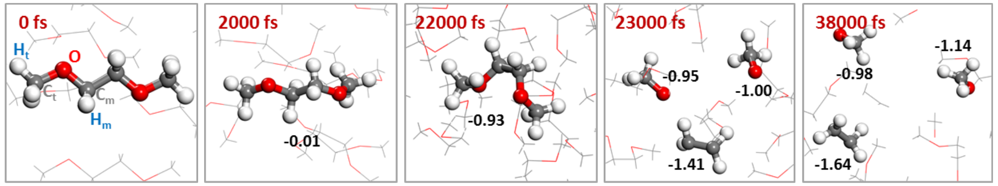
Main Fields where Battery Research is taking Huge Steps

- surface and interfacial characterization, *in situ*, *operando* modes
- **first-principles computations, multiscale methodologies**
- **multimodal characterization (integrating theory and experiments)**
- **data science approaches**

Electrolyte Decomposition under Electron-Rich Environments



Pure DME



$$\mathbf{DME + 4e^- \rightarrow 2CH_3O^- + C_2H_4{}^{2-}} \quad (1.1)$$

$$C_2H_4{}^{2-} + DME + 2e^- \rightarrow 2CH_3O^- + 2C_2H_4{}^- \quad (1.2)$$

$$2C_2H_4{}^- \rightarrow (C_2H_4)_2^{2-} \quad (1.3)$$

1M Salt in DME

$$\mathbf{DME + (frag.)^{x-} \rightarrow DME(-H_t)^- + (H\cdot frag.)^{y-}} \quad \mathbf{(2a.1)}$$

$$\mathbf{DME + (frag.)^{x-} \rightarrow DME(-H_m)^- + (H\cdot frag.)^{y-}} \quad \mathbf{(2b.1)}$$

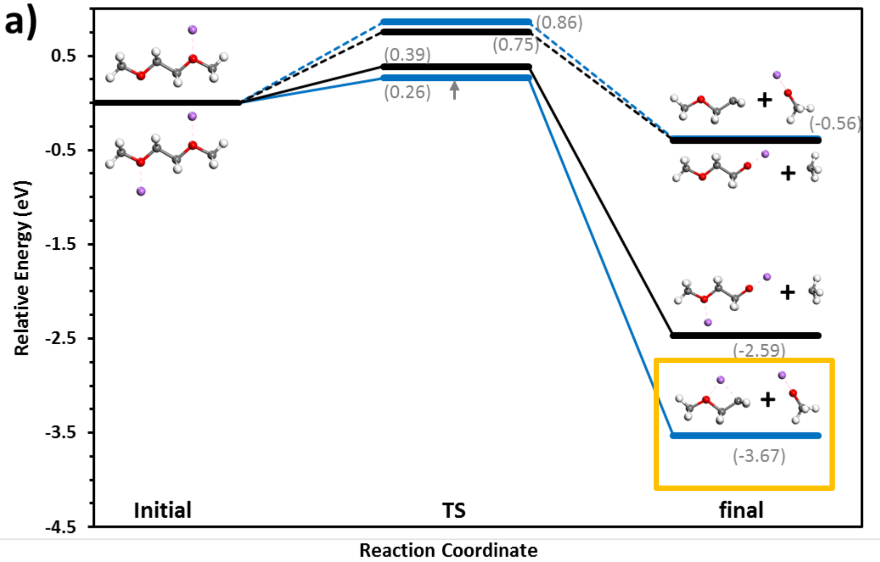
$$DME(-H_m)^- \rightarrow CH_3O^- + CH_3OCHCH_2 \quad (2b.2)$$

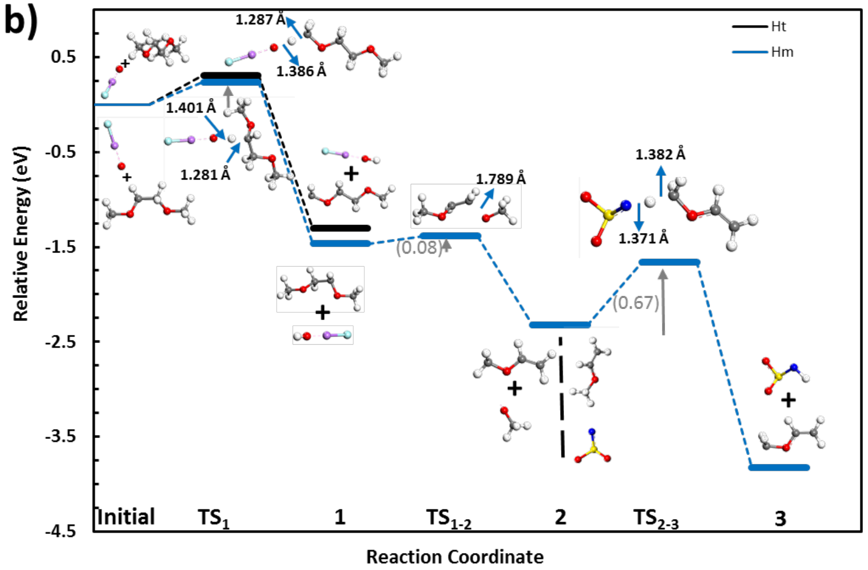
$$CH_3OCHCH_2 + (frag.)^{x-} \rightarrow CH_2OCHCH_2{}^- + (H\cdot frag.)^{y-} \quad (2b.3)$$

Fragments: CSO_2N^{3-}, O^{2-}---LiF_x, $NSO_2{}^{3-}$

○ H ● Li ● F ● O ● S ● N ● C ● – F or – CF_3

Thermodynamics and Kinetics



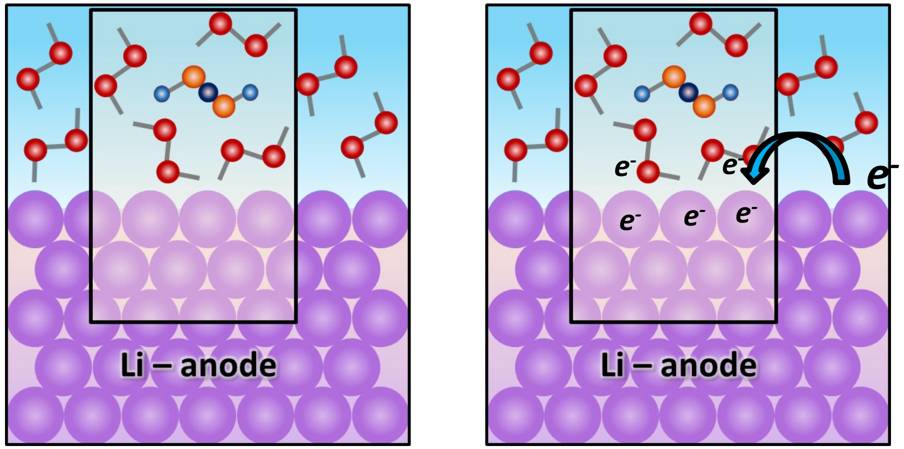


Li-metal/Electrolyte: Effect of External Electrons

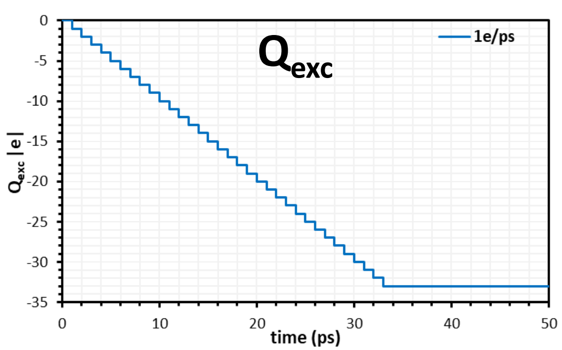
Effect of excess electrons at the Li/Electrolyte Interface:

L.E. Camacho and P. B. Balbuena, JPS 2020

Zero charge *v.* **Q_{exc}**



$Q_{exc}=0$



Luis Camacho's work

Electrolyte Concentrations

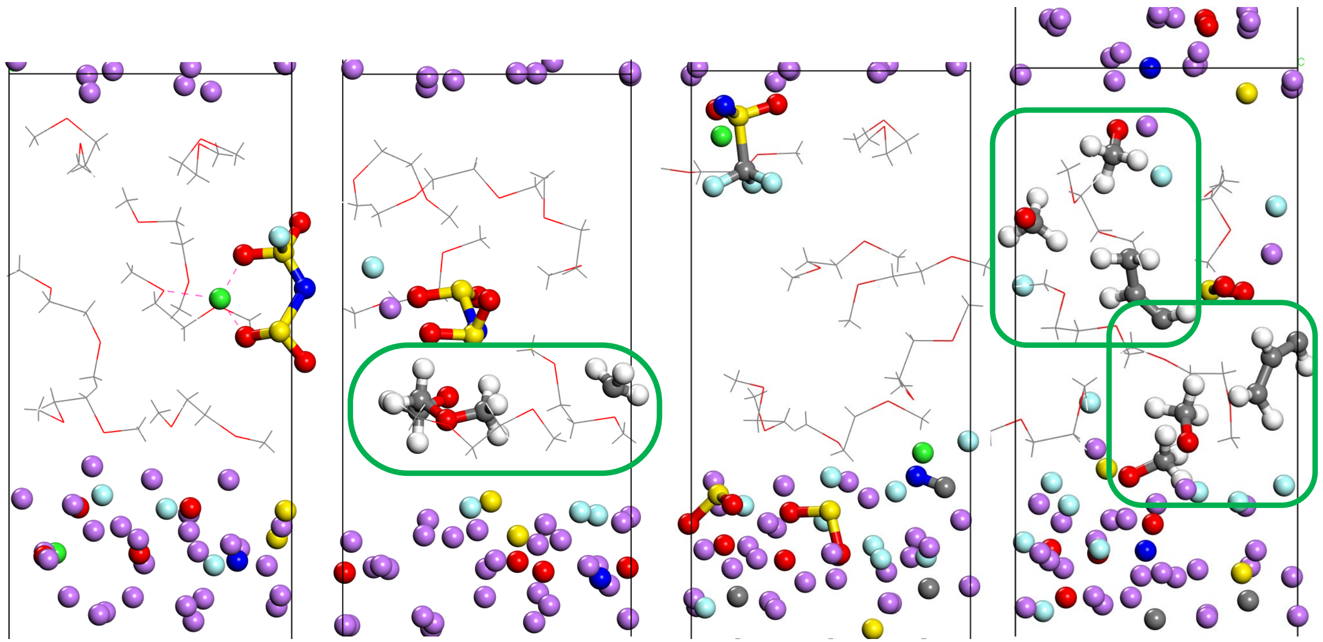
1M and 2M Salt (LiFSI and LiTFSI) in DME

Solvent Only: DME, DOL, and DME:DOL

General Highlights

- **Salt** decomposition occurs w/ and w/o Q_{exc}. However, the extent of decomposition is higher when an excess of charge is added.
- Excess of electrons leads to **solvent decomposition**.
- **Radical anions** formed from solvent or TFSI$^-$ decomposition may **trigger** additional DME or DOL **decomposition**.

Zero charge | Q_{exc} | Zero charge | Q_{exc}

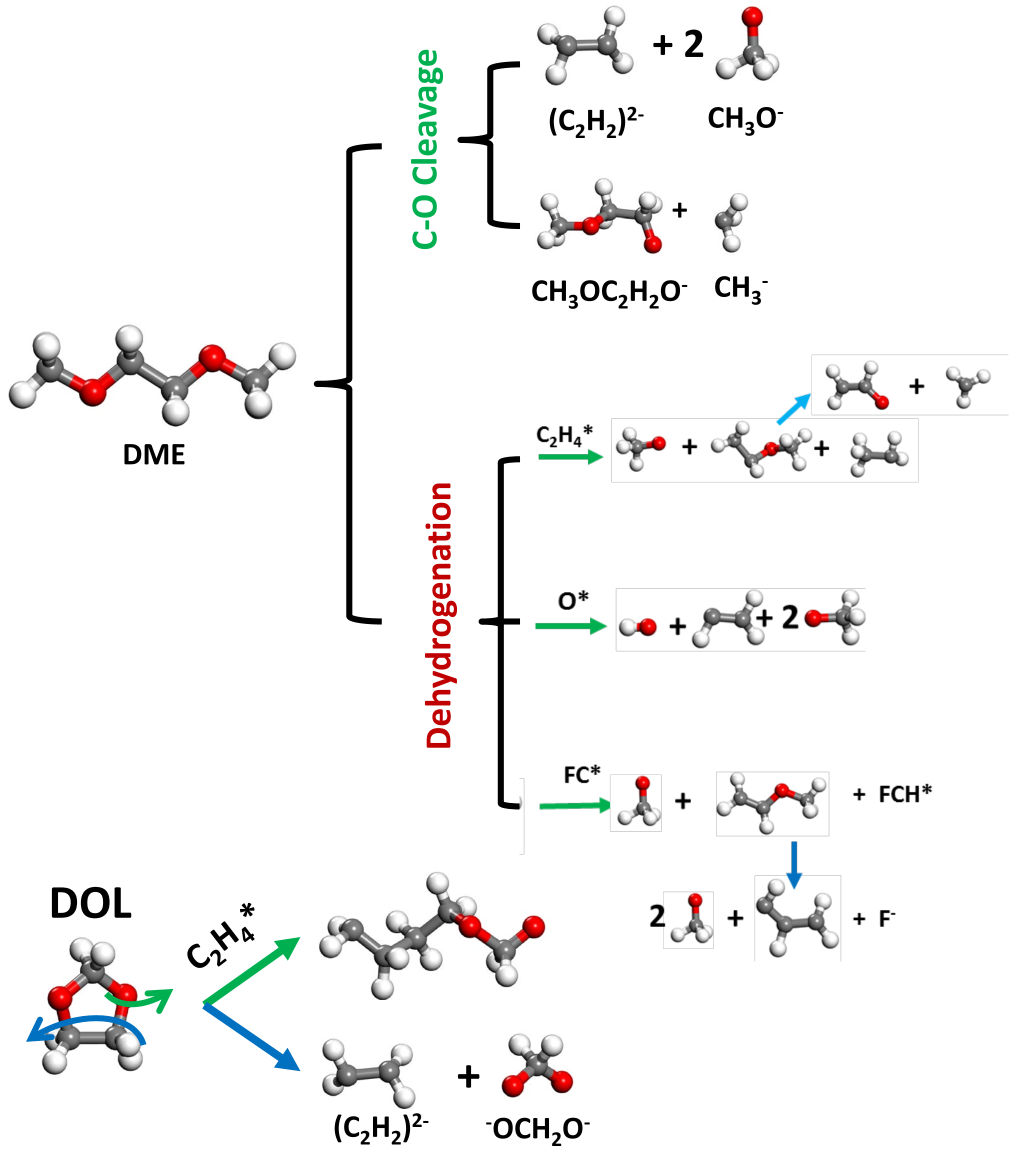


2M-LiFSI | **2M-LiTFSI**

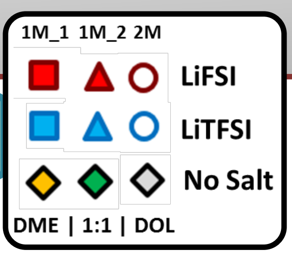
H | Li | F | O | S | N | C

Li-metal/Electrolyte: Effect of External Electrons

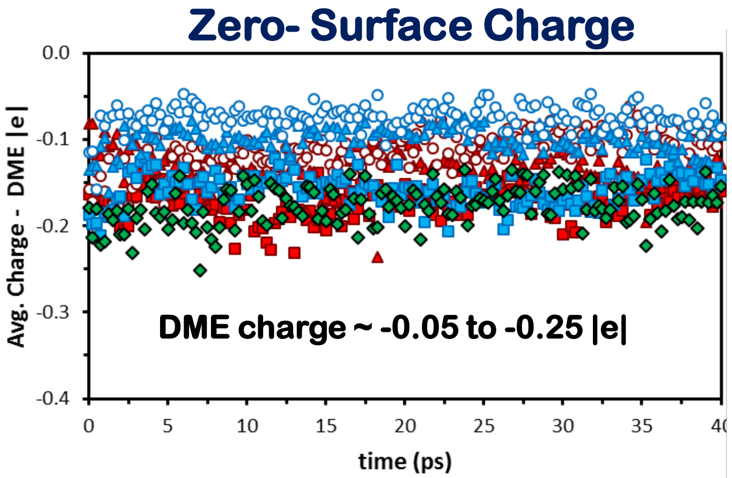
Solvent Decomposition

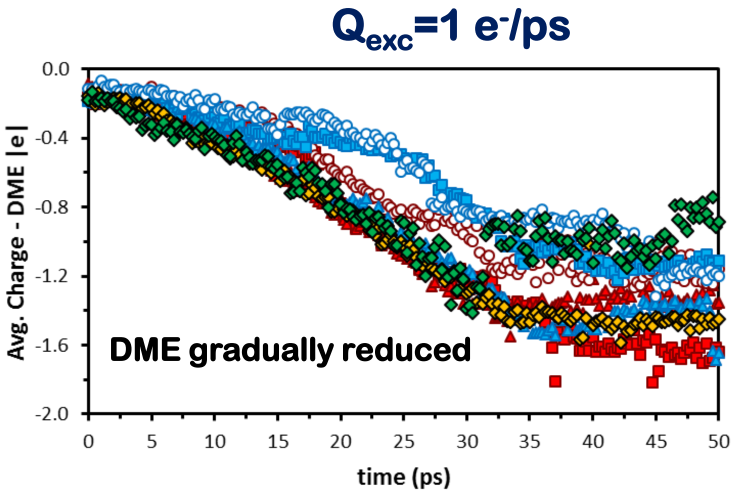


Overall Charge Analysis

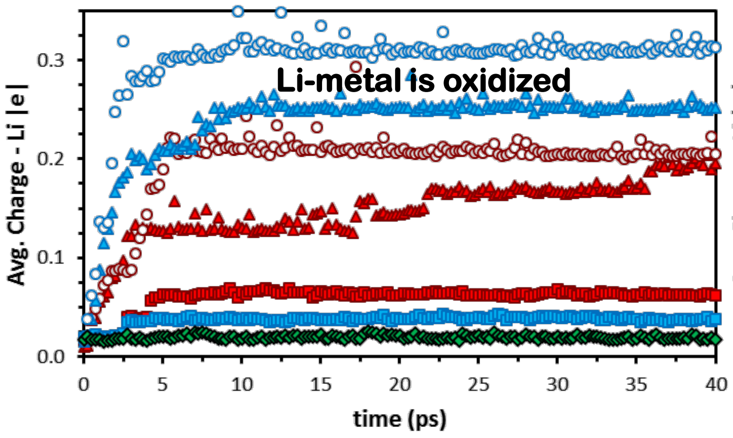


Avg. Charge – DME

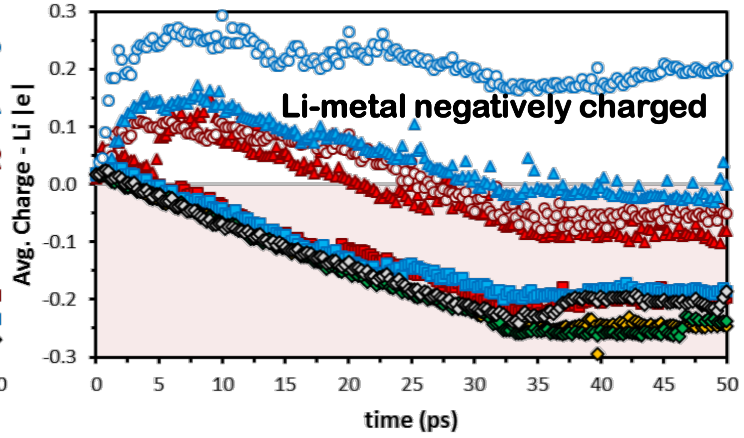




Avg. Charge – Li (slab)



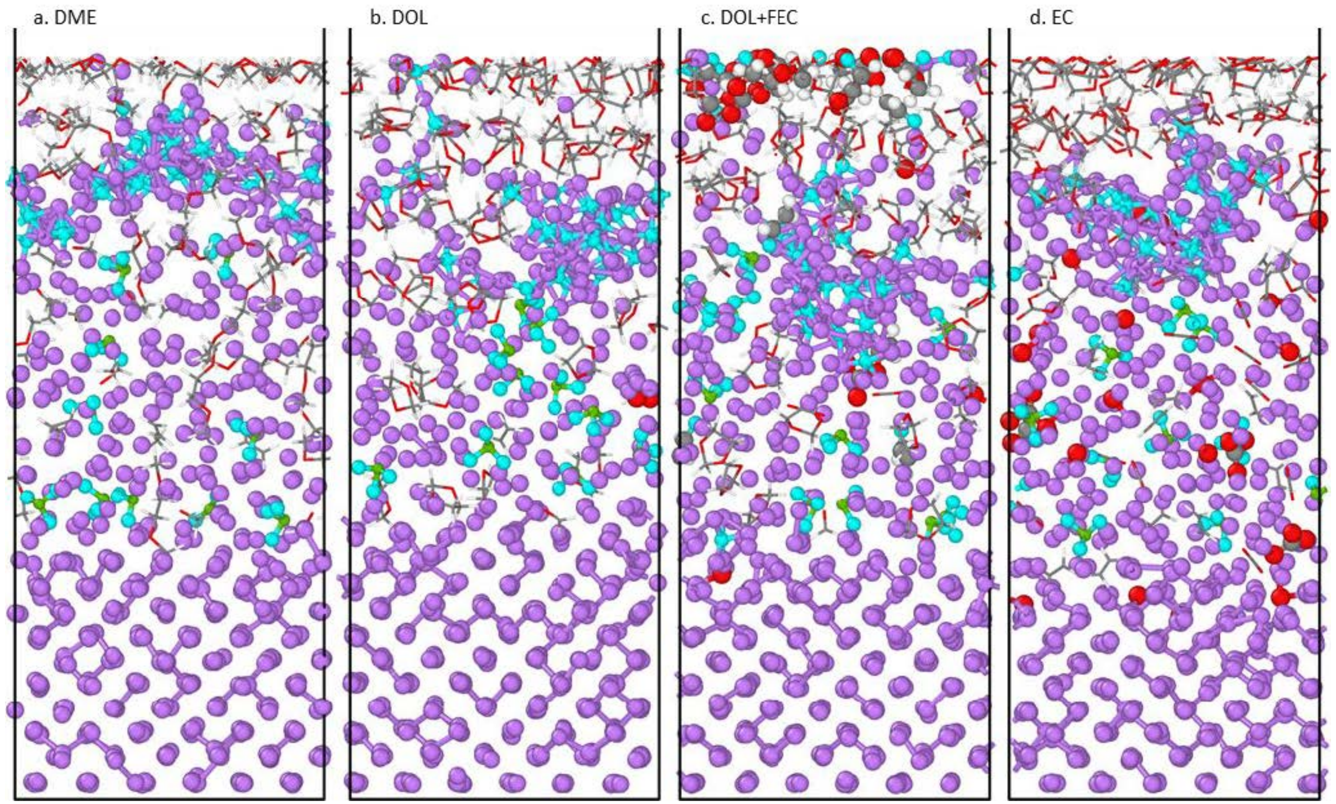
Li oxidation → pulverization



H Li F O S N C

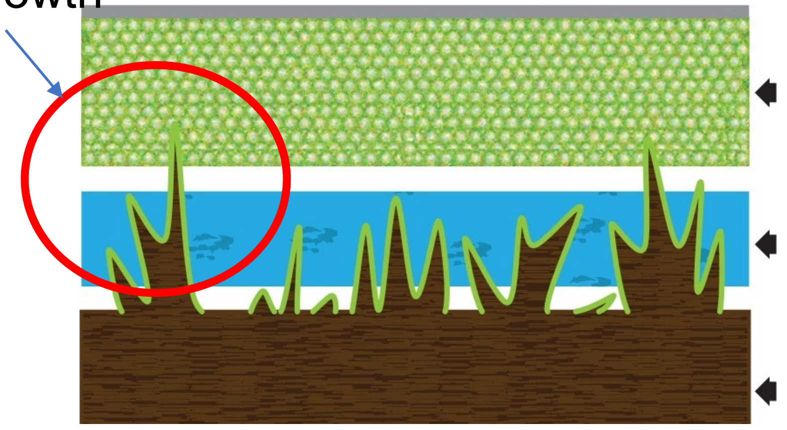
Reactive molecular dynamics: Passivation layers at the Li anode surface

Ospina-Acevedo, Guo, Balbuena, J. Mater Chem A, 2020

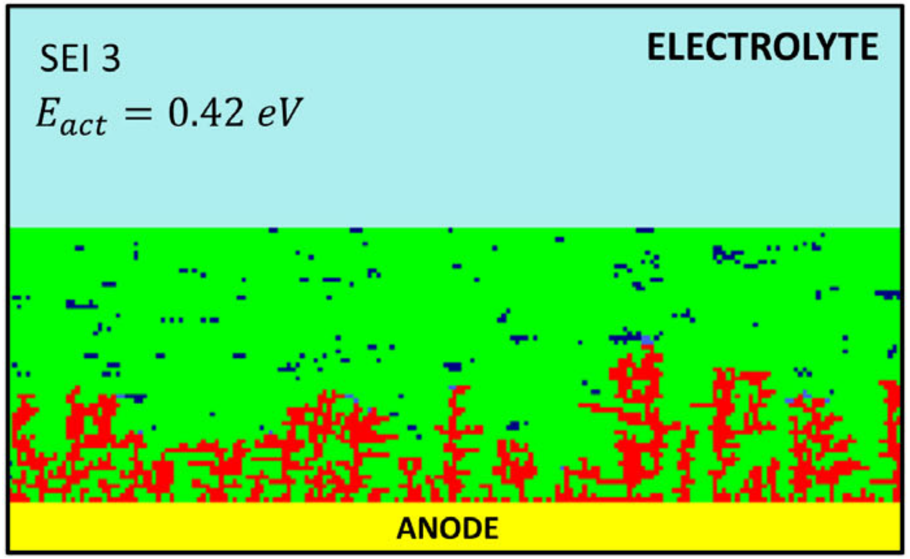


Li metal dendritic growth

Schematic during Li Plating



$\eta = 0.2\ V;\ T = 300\ K$



MD + KMC simulations
Sitapure, Lee, Ospina-Acevedo, Balbuena, Hwang, Kwon, AIChE J. 2020

Concept of Nanobattery (Seminario's group, CHEN)

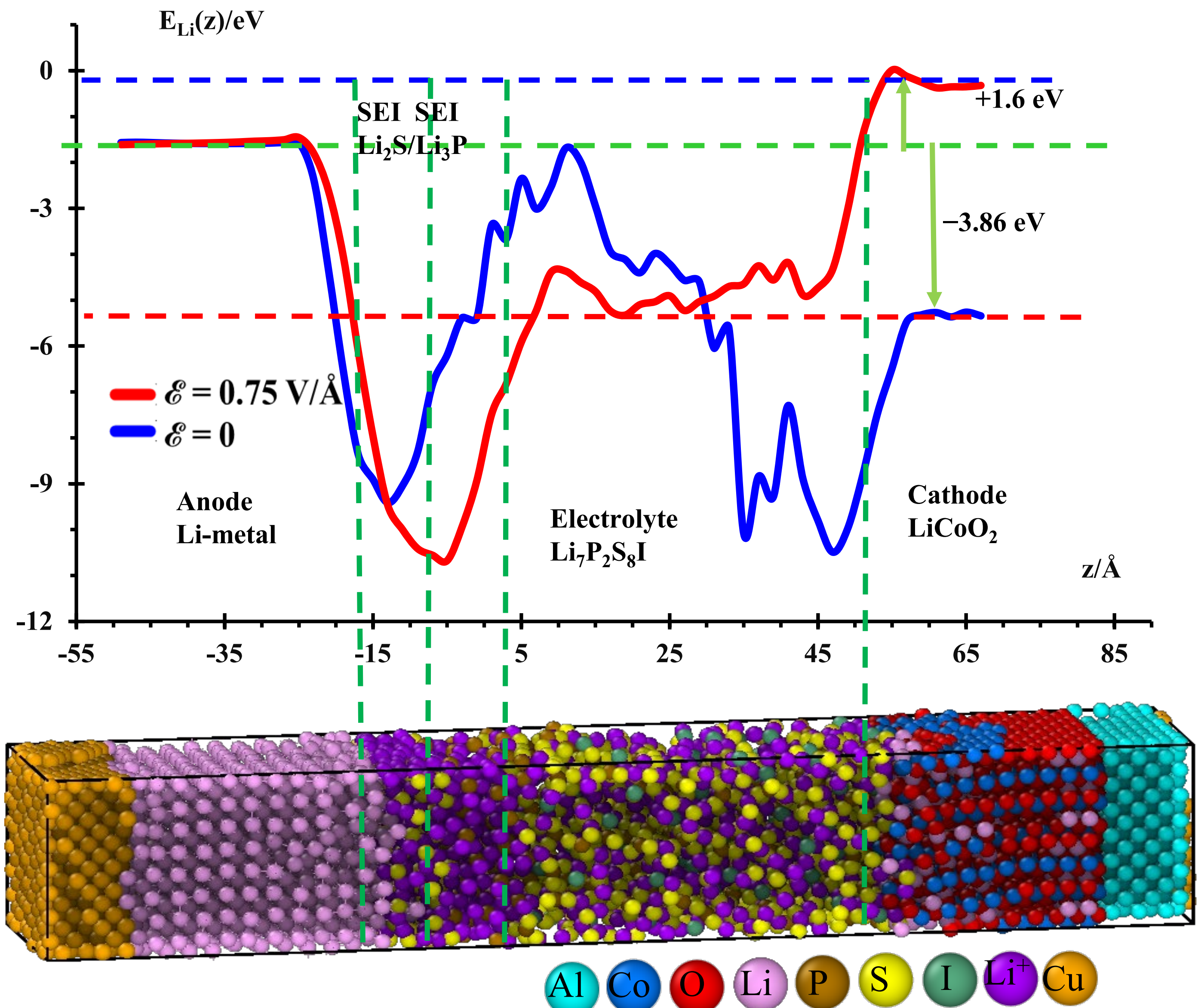
Classical MD simulations of solid-state electrolyte $Li_7P_2S_8I$

Ponce, Galvez-Aranda, Seminario; *PCCP*, *23*, 597-606, **2021**

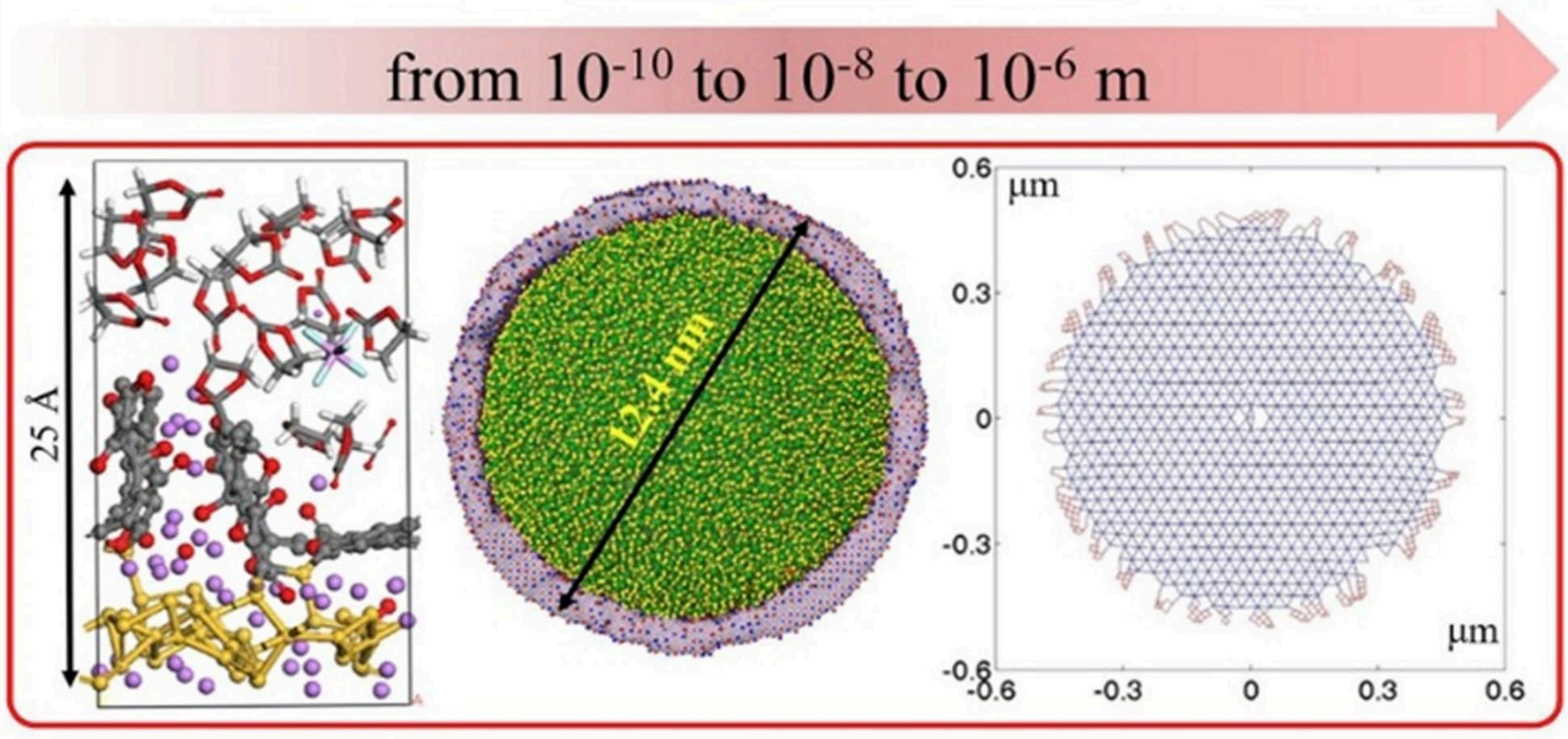
average energy profile experienced by Li ion as it travels from cathode to anode

estimate of the overpotential needed to overcome the energy difference between cathode and anode during charge

Li energy average w/ & without EF



Chemical + Mechanical Properties: Multiscale Modeling Si Anodes



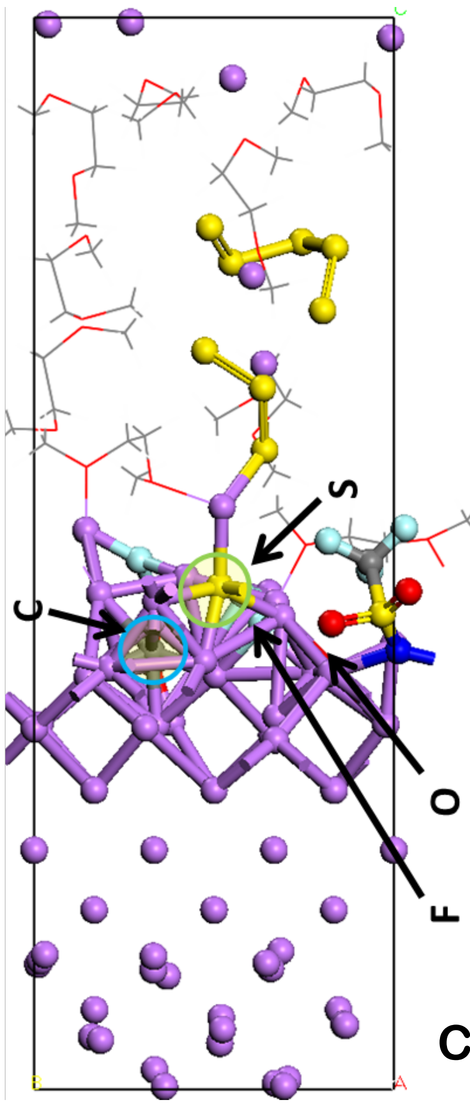
surface/electrolyte interactions at atomistic, nanoscale, mesoscopic levels

D. E. Galvez-Aranda, A. Varma, K. Hankins, J. M. Seminario, P. P. Mukherjee, and P. B. Balbuena, "Chemical and Mechanical Degradation and Mitigation Strategies for Si Anodes", J. Power Sources, 419, 208-218, (2019).

Multimodal Characterization of Passivation Layers

Solvent: DME and DOL

AIMD and *In Situ* XPS Imaging



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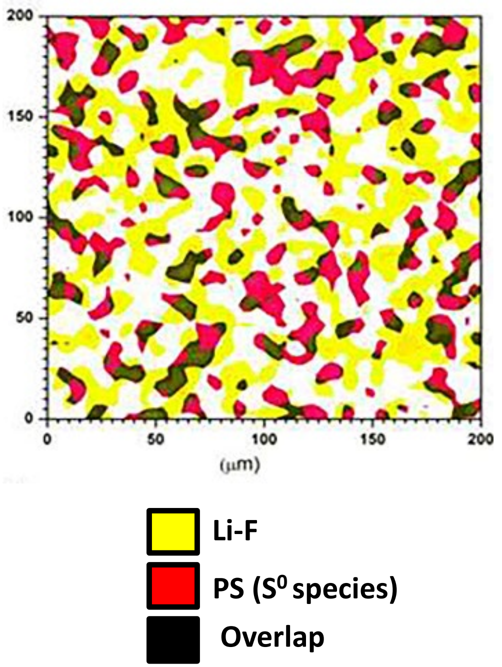
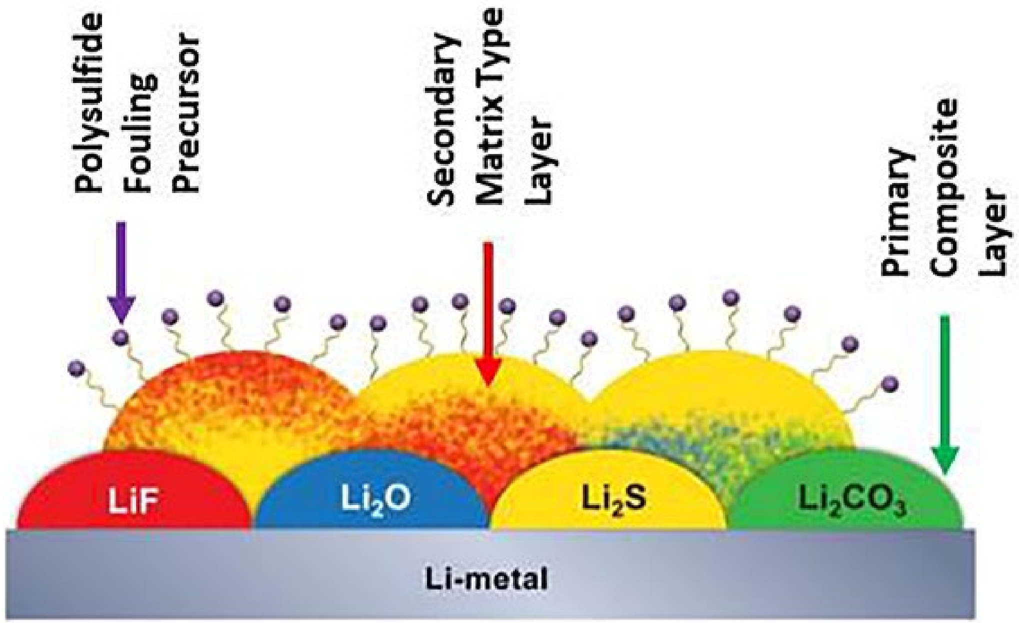


SEI Layer Growth



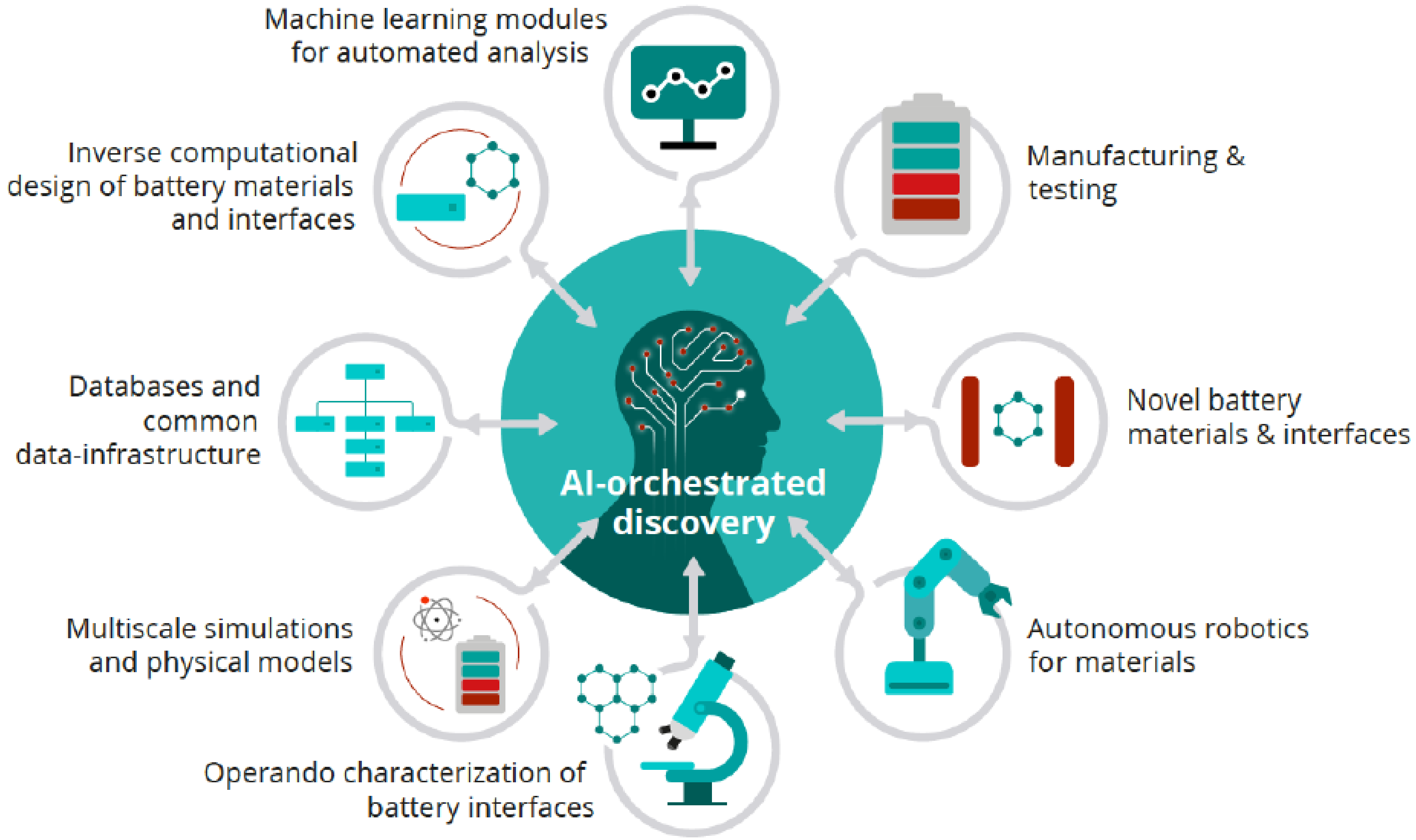
Collaboration with Vijay Murugesan, PNNL

Chem. Mater., **2017**, 29 (11), 4728–4737

Important point:
presence of SEI blocks that are
not mono-components
but multi-components

H Li F O S N C

Artificial Intelligence: Promise of Further Discovery



From Battery 2030 Road Map, European Union, 2020

Conclusions

- High-performance computing: crucial role in the understanding and design of materials for a highly interactive system
- Quantum and classical molecular dynamics, multiscale modeling approaches
- Analysis of localized and integrated systems
- Predictions and integration with experiments
- Data science approaches

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