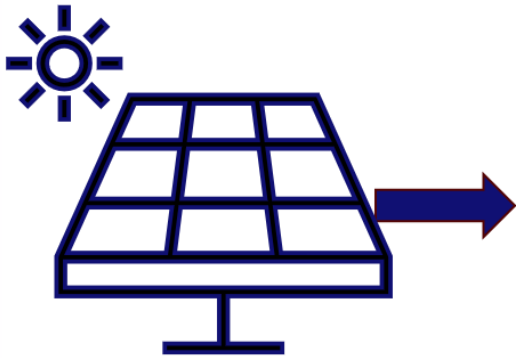
Interface engineering for energy conversion and storage technologies

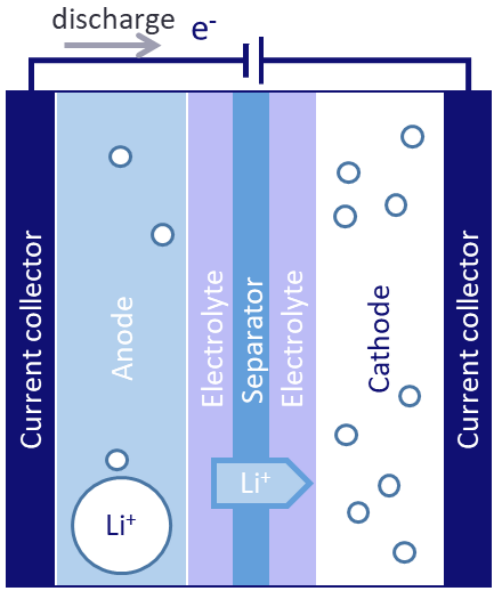
Adriana Creatore

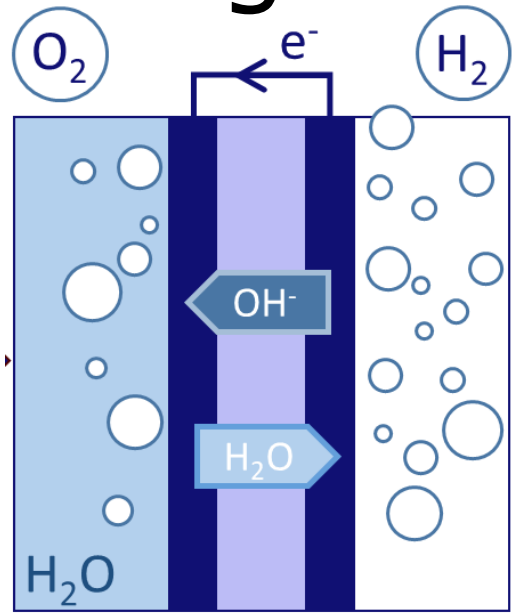
Applied Physics and Science Education department & Eindhoven Institute of Renewable Energy Systems (EIRES), TU/e

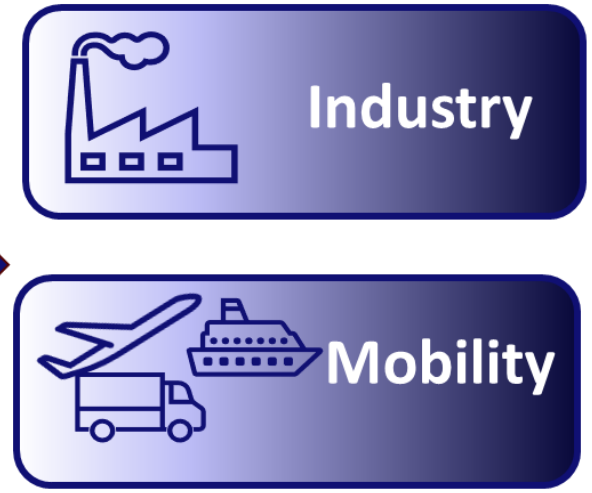


Future energy technologies









Photovoltaics, beyond thermodynamic limit of single junction PV

High energy density and safe Li- ion batteries

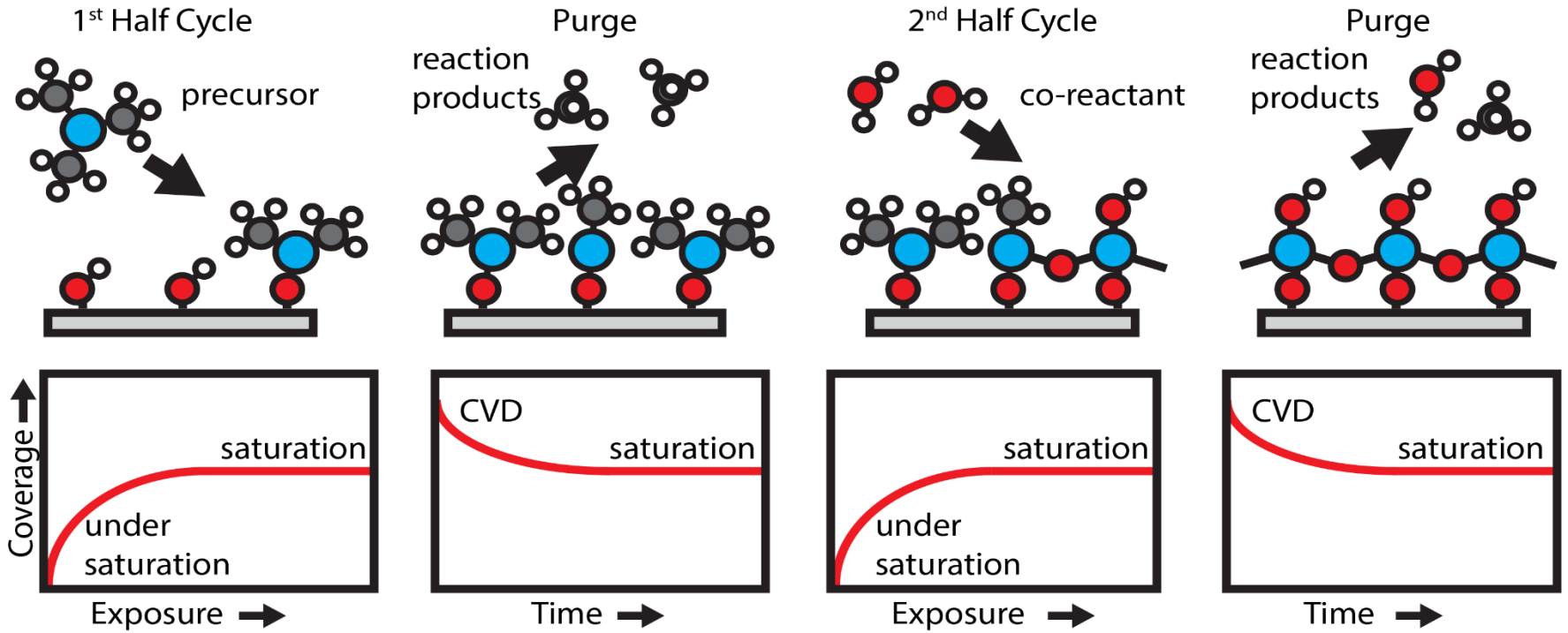
H_2O electrolysis, low CRMs

Electrification of mobility & chemical/manufacturing industry

Scale mismatch:
performance and lifetime of energy devices governed by interfaces

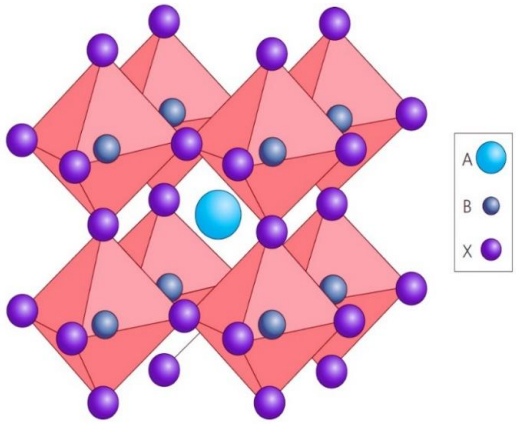
Atomic layer deposition (ALD)

An ALD cycle consists of two (or more) “half-reactions” separated by purge and/or pump steps. The “half-reactions” are self-limiting leading to well-defined amount of added material. Every step of the ALD process takes place in “saturation”.



Ångström-level growth control- excellent conformality - unparalleled uniformity

Metal halide perovskite PV

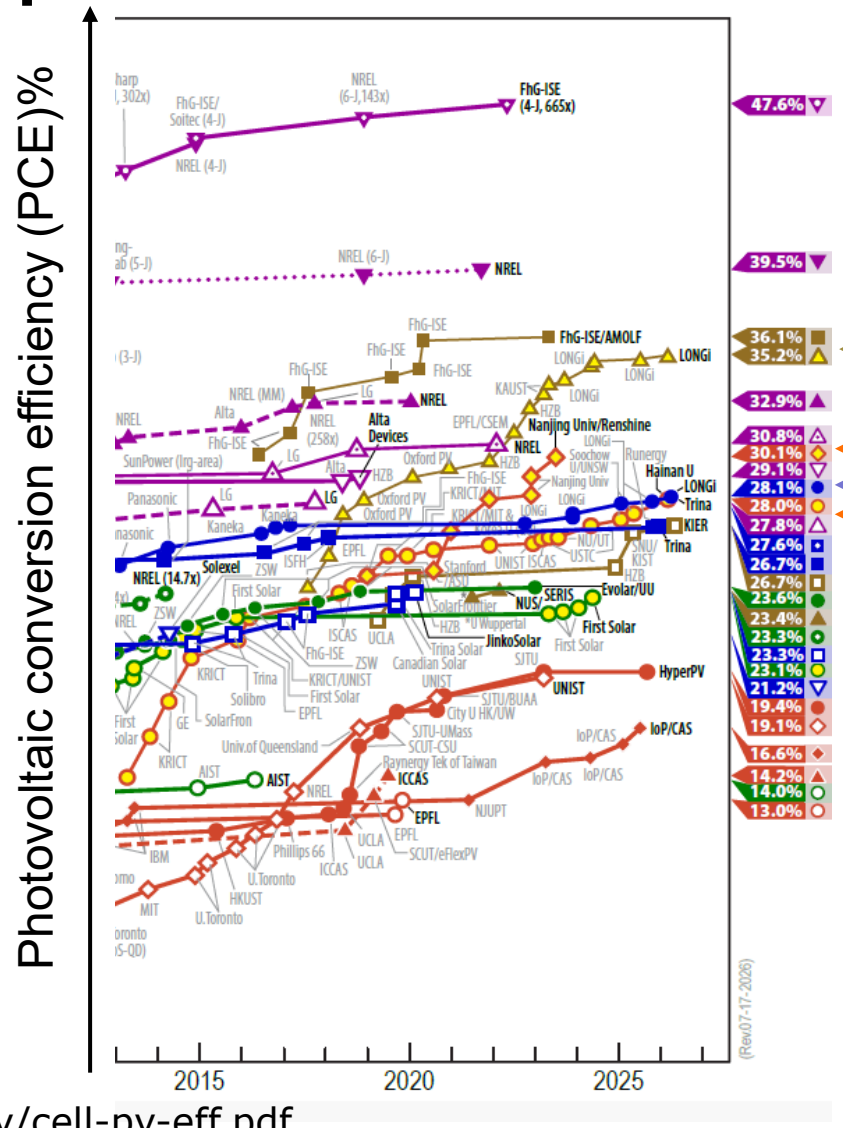


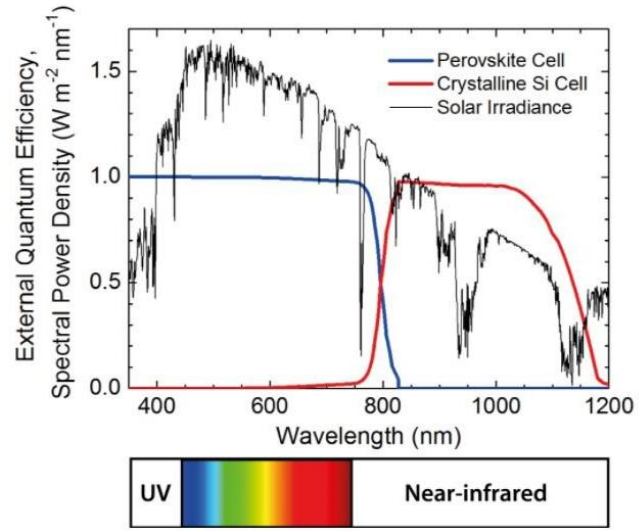
Perovskite crystal structure, ABX_3

Cation ($CH_3NH_3^+$, $HC(NH_2)_2^+$, Cs^+)

Metal cation (Pb^{2+},Sn^{2+})

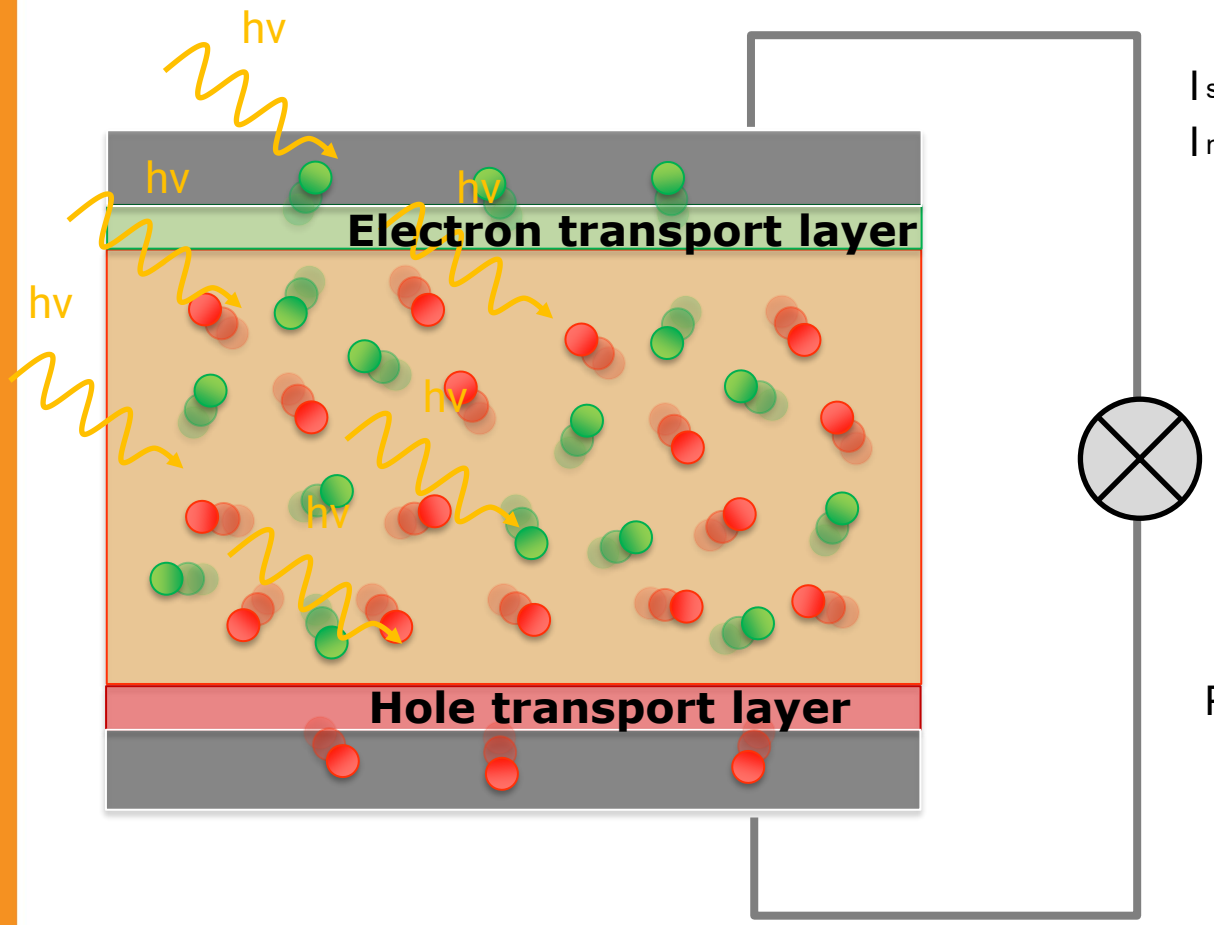
Halide anion (I^-,Br^-,Cl^-)



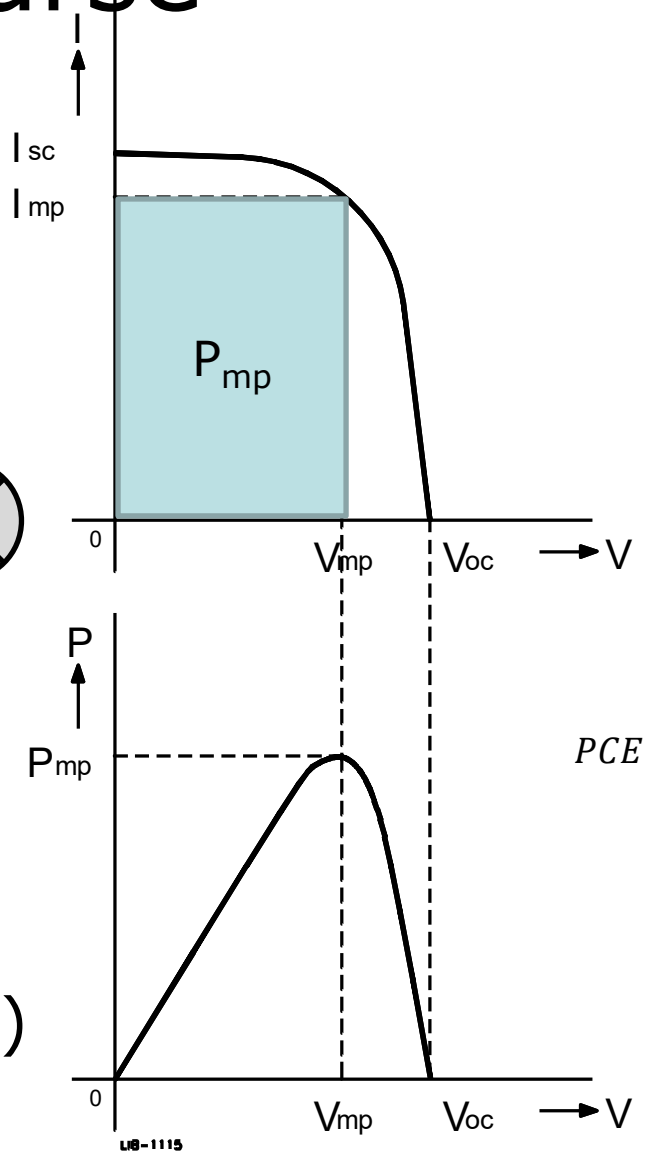


https://www.nlr.gov/docs/libraries/pv/cell-pv-eff.pdf

A solar cell crash course



Semiconductor (c-Si, perovskite…)



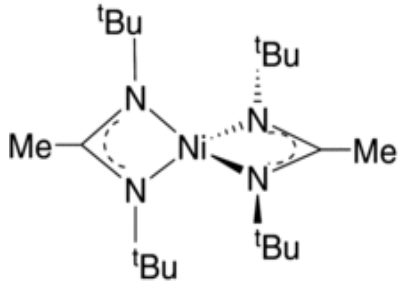
$$FF = \frac{V_{mp} \times I_{mp}}{V_{oc} \times I_{sc}}$$

$$P_{mp} = FF \times V_{oc} \times I_{sc}$$

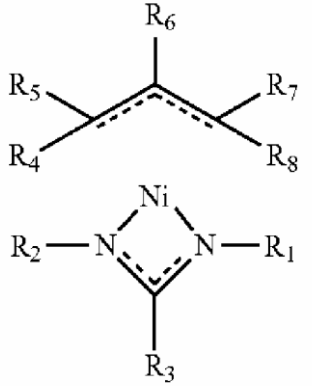
$$PCE = \frac{\mathrm{P_{mp}(electric\ out)}}{\mathrm{P_{opt}(optical\ in,\ whole\ spectrum)}}$$

ALD NiO hole transport layers

Cu
Electron transport layer
Perovskite absorber $CsFAMAPbI_3$
Hole transport layer (NiO)
TCO
Glass

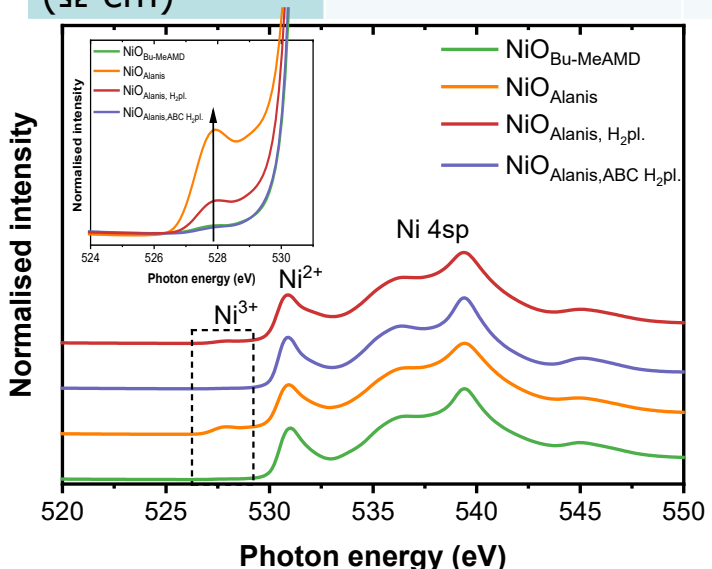


$Ni(^tBu\text{-}MeAMD)_2 + H_2O$



Alanis + O_2 plasma

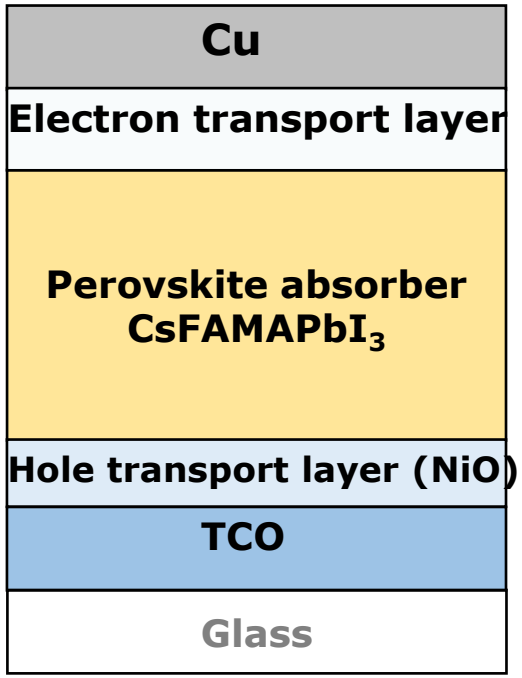
	$NiO_{Bu\text{-}MeAMD}$	NiO_{Alanis}	$NiO_{Alanis,H2\ pl.}$	$NiO_{Alanis,ABC\ H2\ pl.}$
Ni : O	0.93 ± 0.05	0.86 ± 0.05	0.85 ± 0.05	0.94 ± 0.05
Resistivity (Ω·cm)	$(1.9 \pm 0.3)\cdot10^4$	4 ± 1	$(1.1 \pm 0.1)\cdot10^4$	Beyond measurement limit

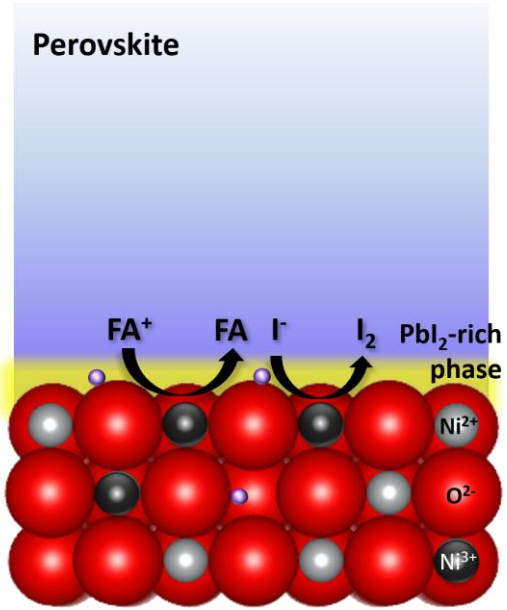


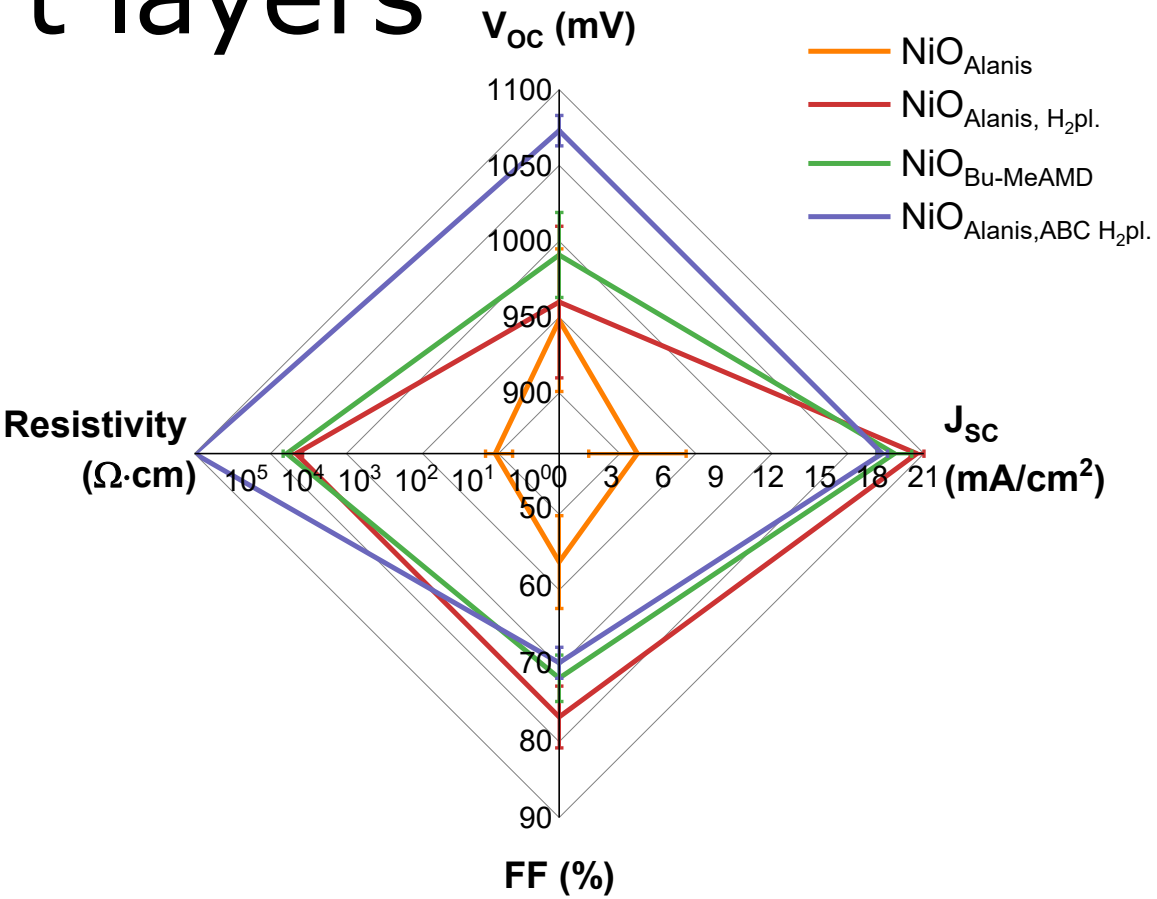
Wide range of NiO bulk properties achieved via ALD: Ni^{3+} states, responsible for NiO conductivity

Mukherjee *et al.*, ACS Appl. Energy Mat., 2026

ALD NiO hole transport layers



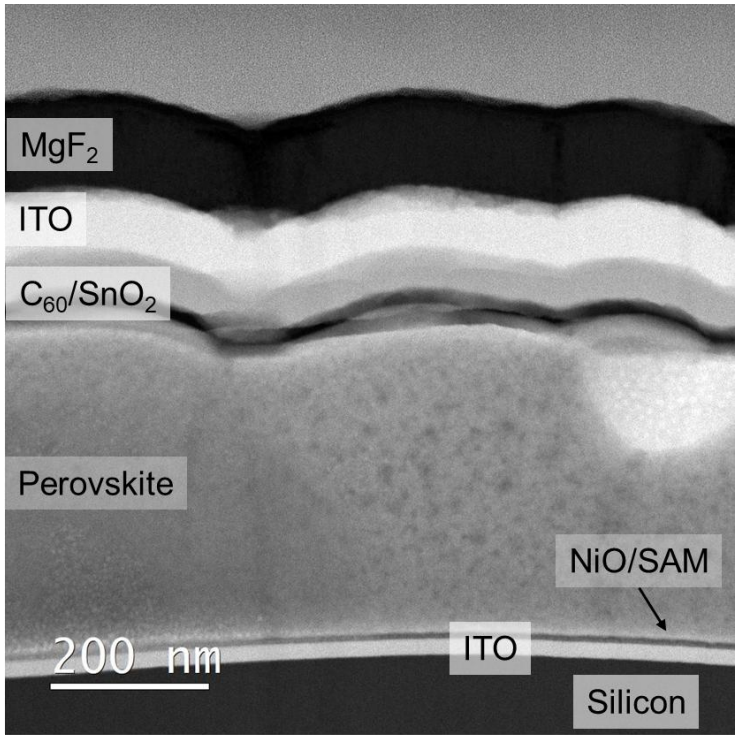


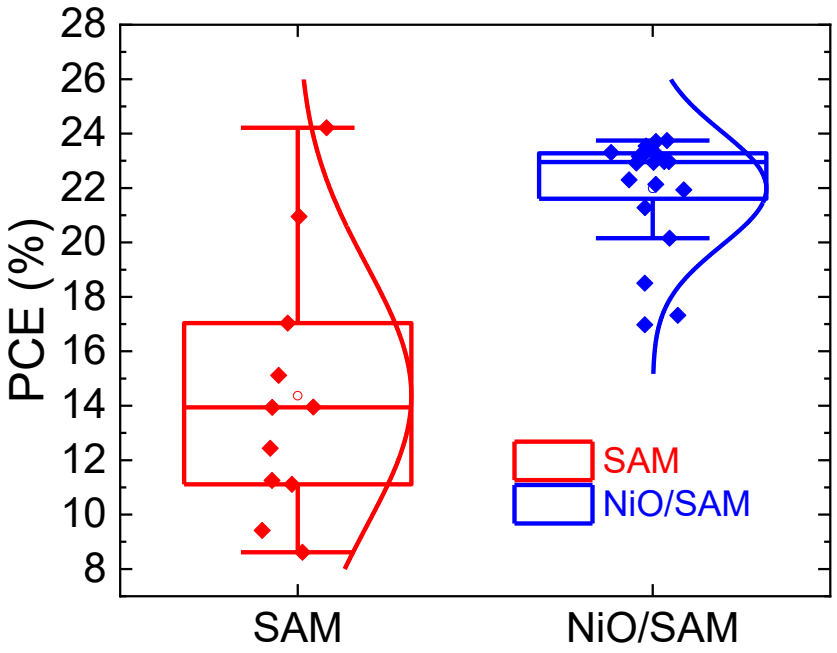


Redox reactions between Ni^{3+} states and perovskite: charge extraction barrier, poor device efficiency.

H_2 plasma suppresses Ni^{3+} states, (J_{sc}, $V_{oc}\uparrow$). However, high resistivity affects charge transport through NiO (FF≈ 75%). Adopt lower NiO thickness (< 8 nm).

ALD NiO in tandem PV

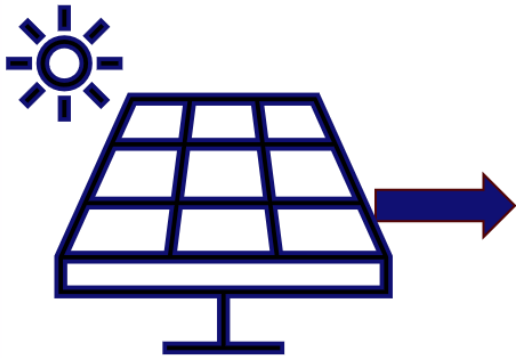


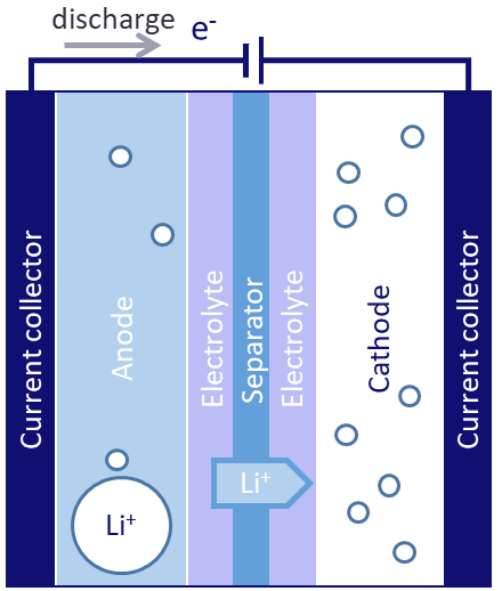


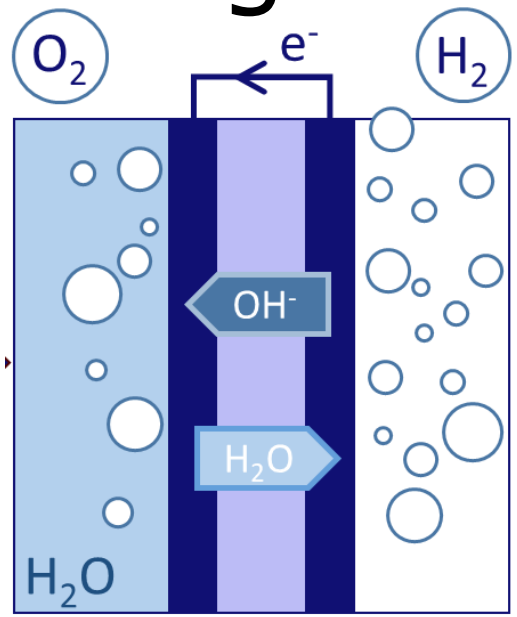
Increased device yield for NiO/SAM in tunnel recombination junction. Tunnel junction ITO/NiO/SAM: almost "lossless" V_{oc} of 1.71 V ($V_{oc,\ Si}$=0.60 V, $V_{oc,\ perovskite}$=1.16 V)

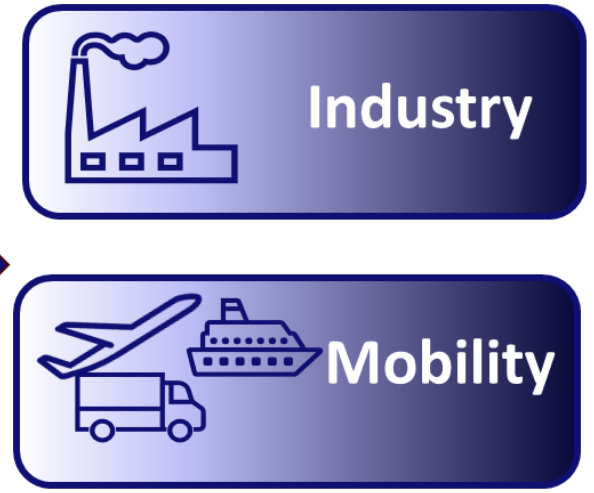
Phung *et al., Sol. En. Mat. Solar Cells* 261, 112498, 2023

Future energy technologies









Photovoltaics, beyond thermodynamic limit of single junction PV

High energy density and safe Li- ion batteries

H_2O electrolysis, low CRMs

Electrification of mobility & chemical/manufacturing industry

Next-generation Li-ion batteries

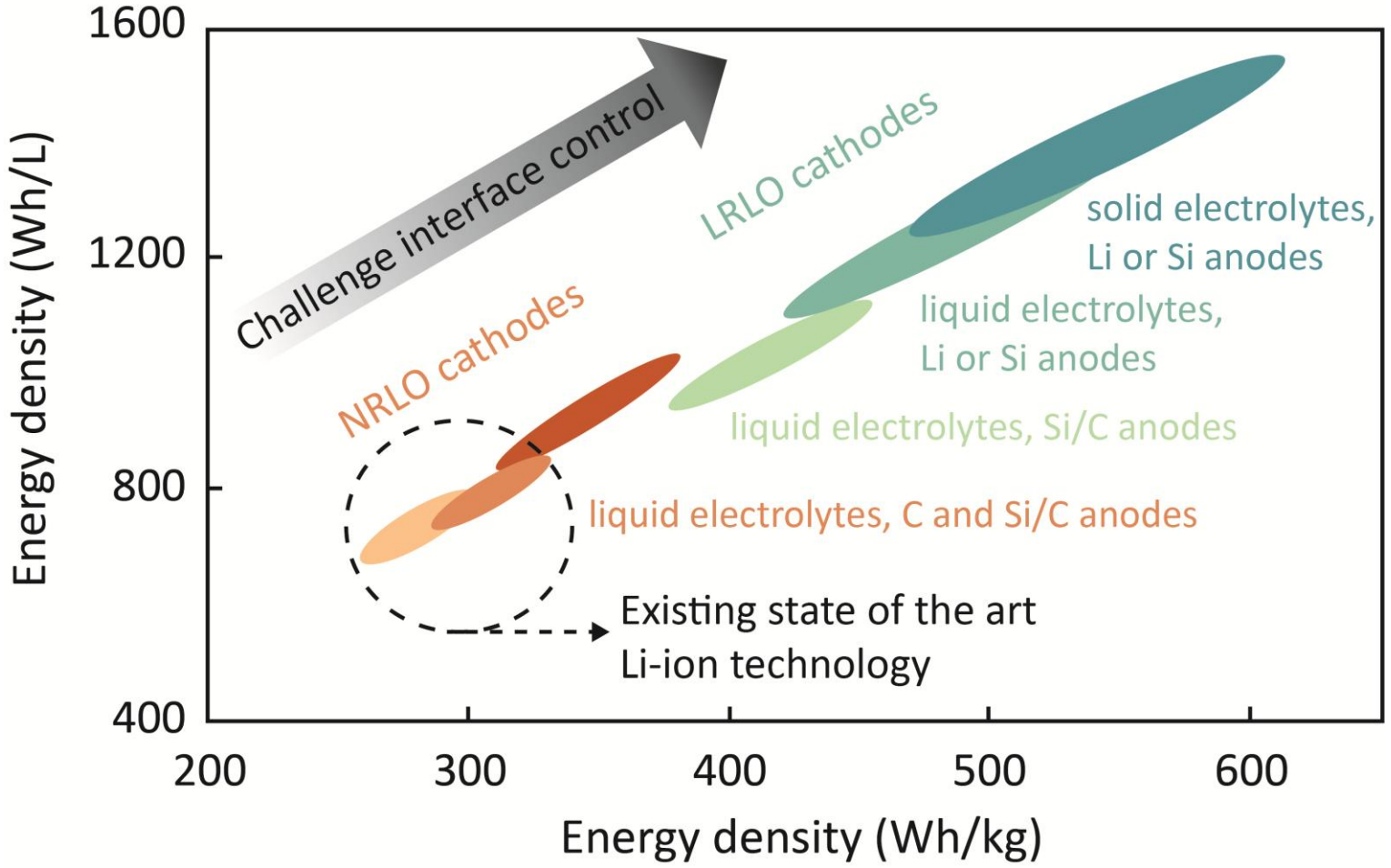


Figure adapted from Schmuch *et al., Nature Energy* 3, 267, 2018

ALD for next-generation Li-ion batteries

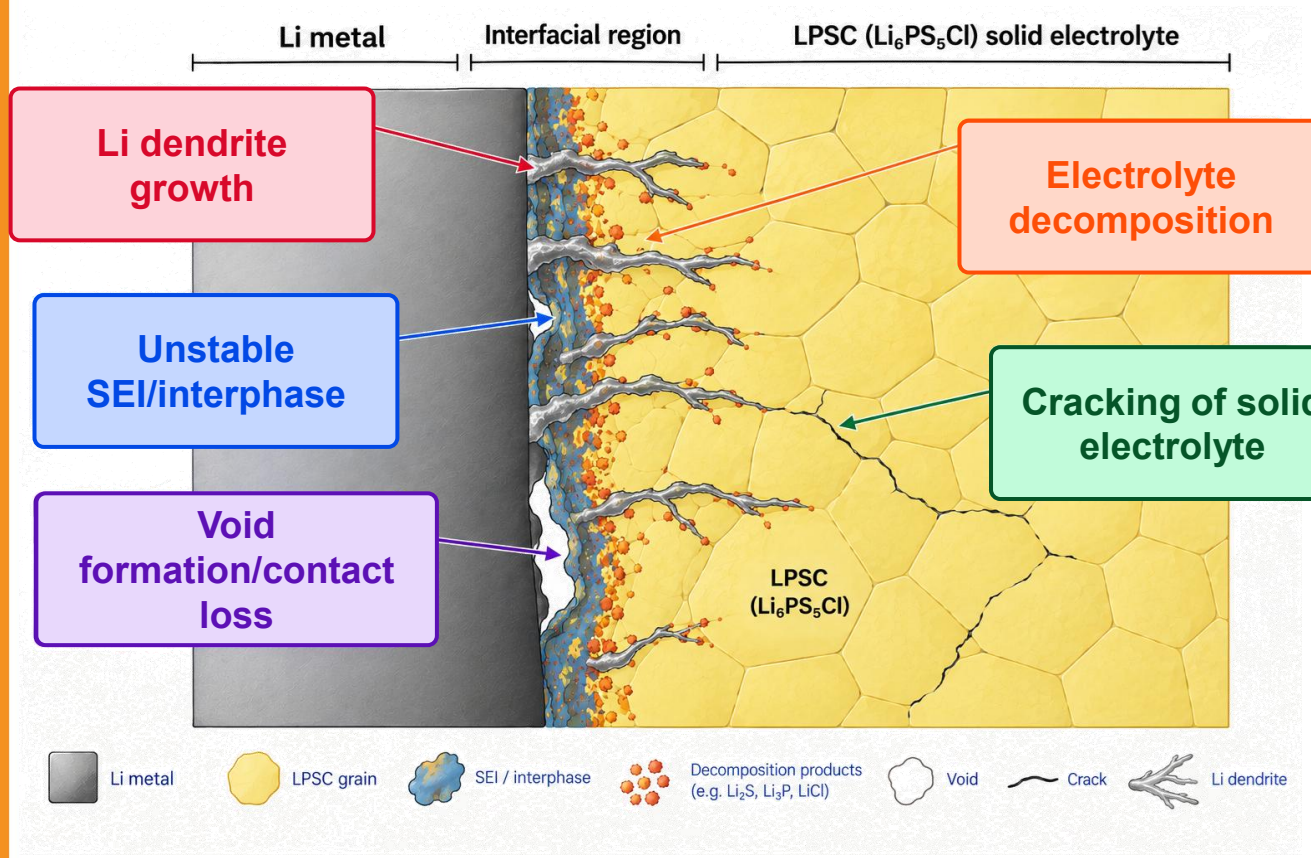
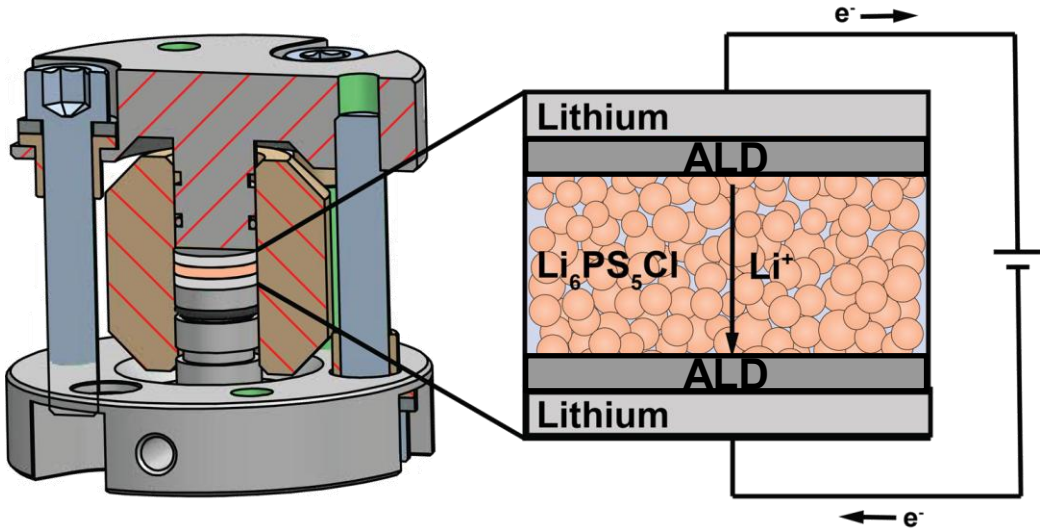
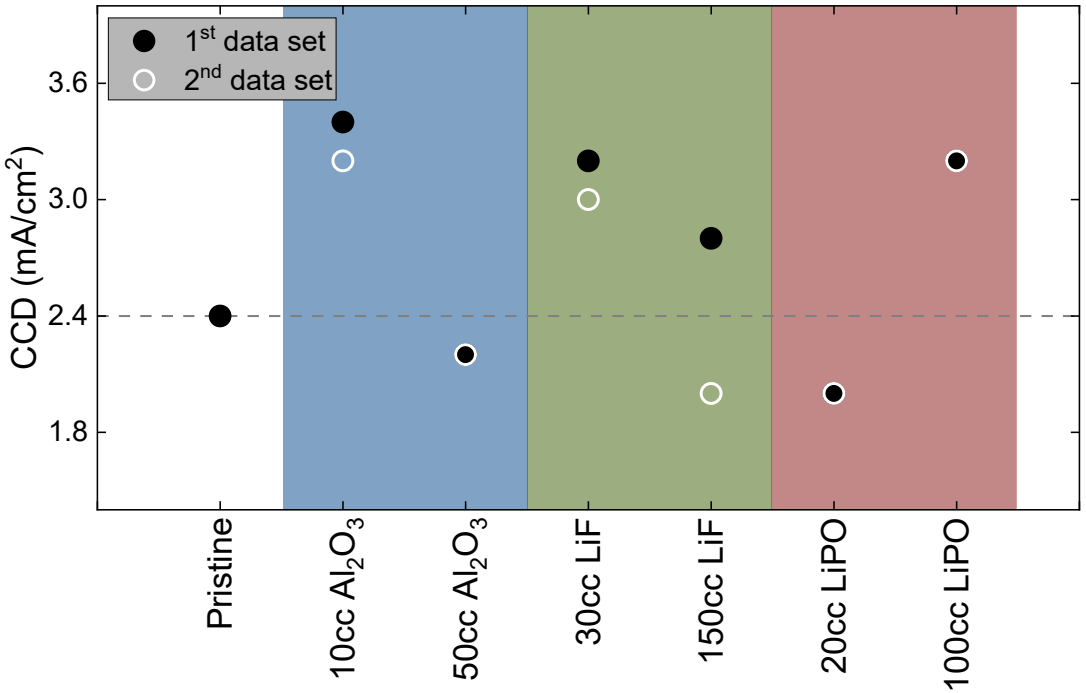


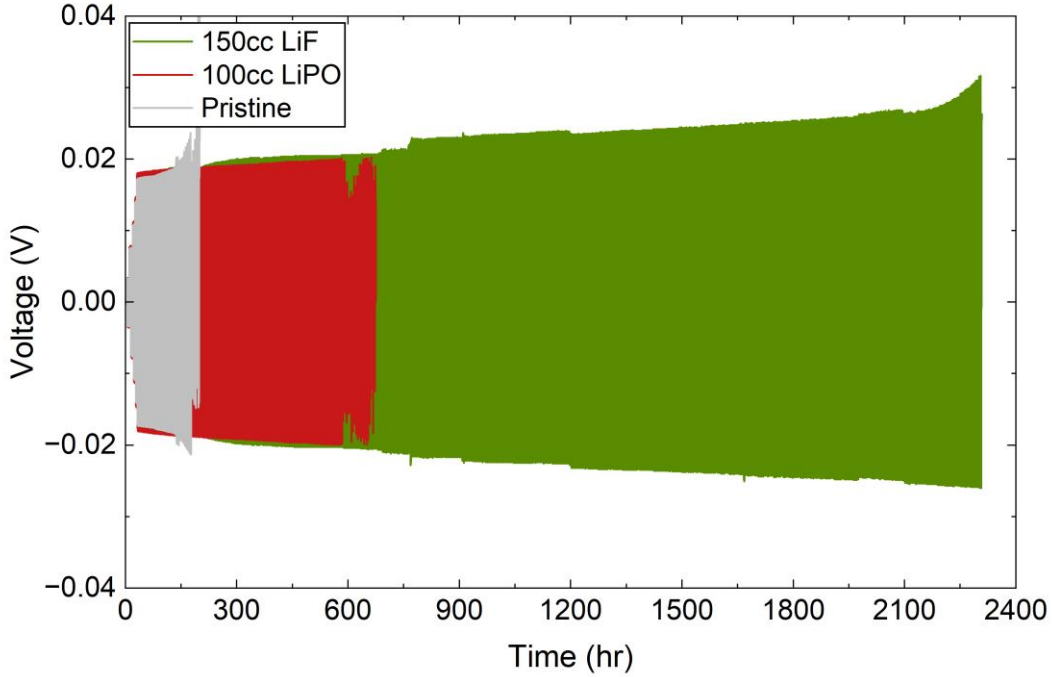
Image generated by ChatGPT



Screening of ALD chemistries (Al_2O_3, LiPO, LiF) by electrochemical testing

Critical current density & long-term cycling



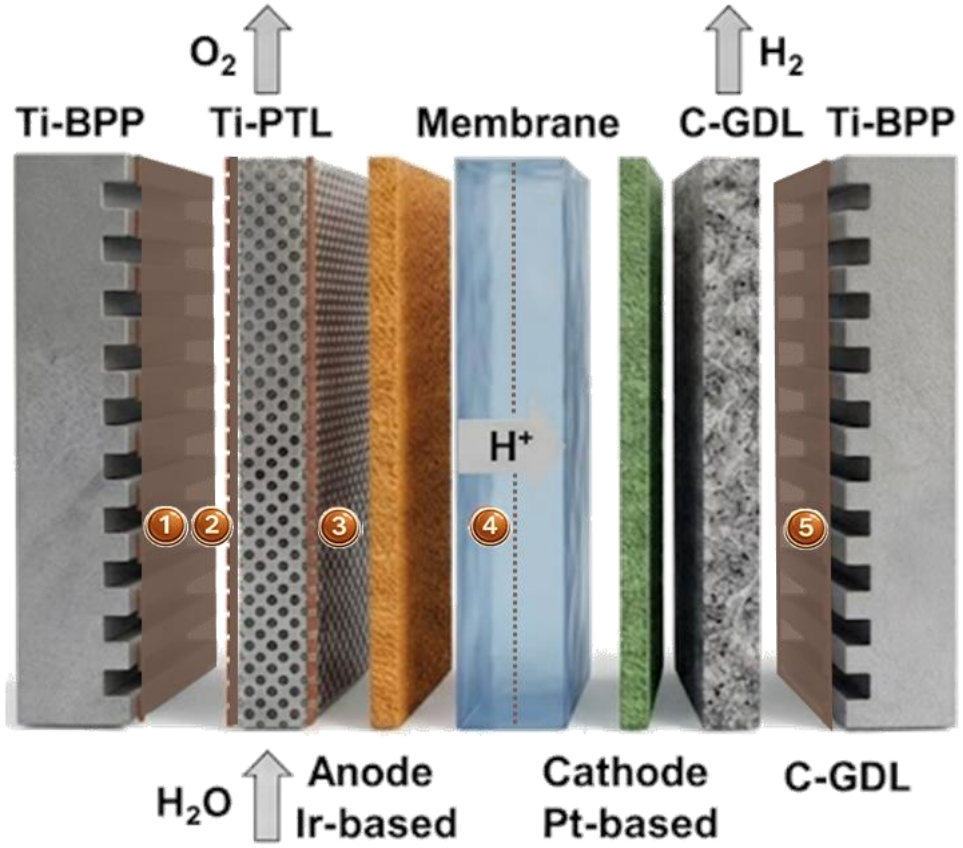


Larger **critical current density** with respect to pristine Li/electrolyte interface

Cause for improvement: suppression of electrolyte decomposition products; increase the Li^+ conductivity at the interface

Cycled at 1 mA/cm^2, Increase in **cycling stability** for ALD **LiF**. Post-mortem analysis necessary to gain insights into role of ALD layer on cell stability

ALD for (PEM) H_2O electrolysis

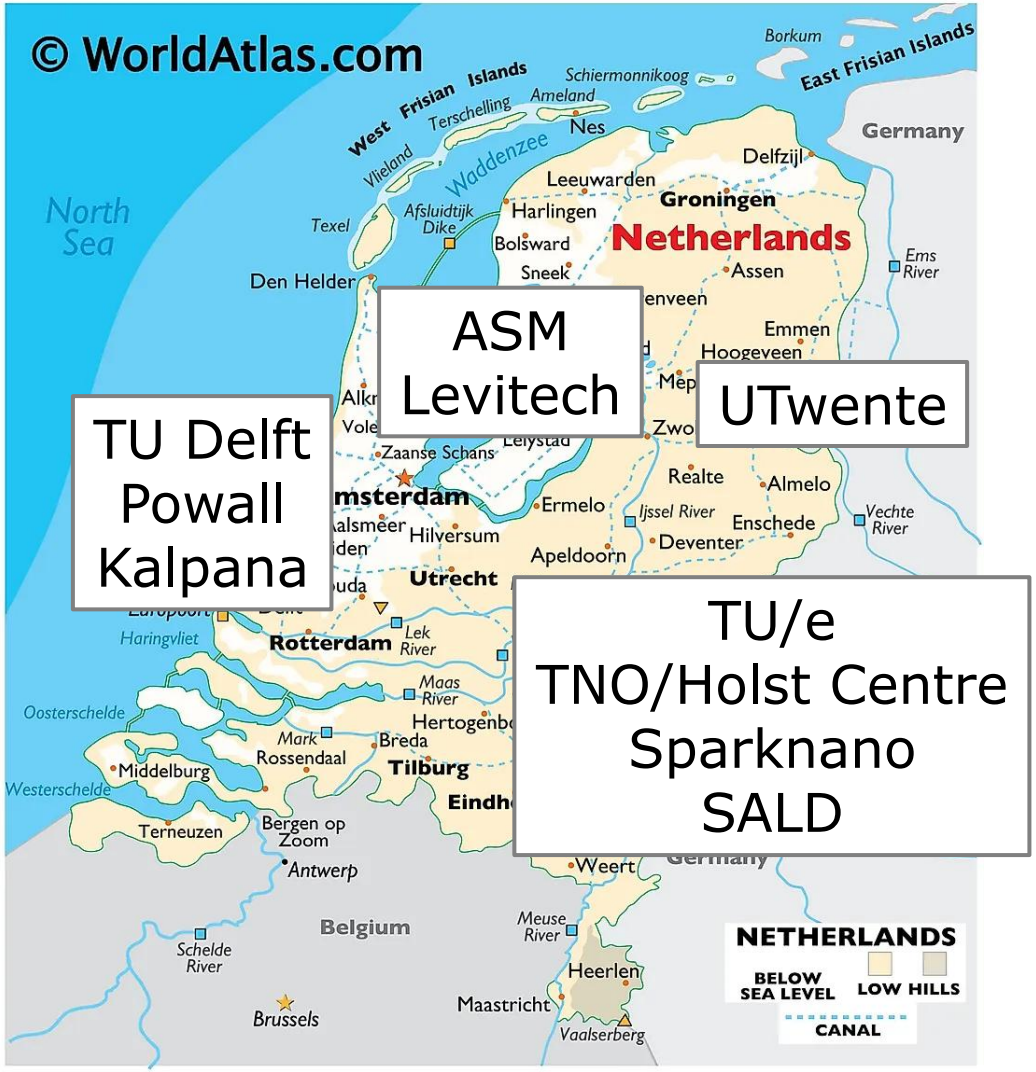


CRMs distributed across the whole PEM stack. Objective: CRM reduction without compromising performance (≥ 3A/cm² for more than 300 hrs)

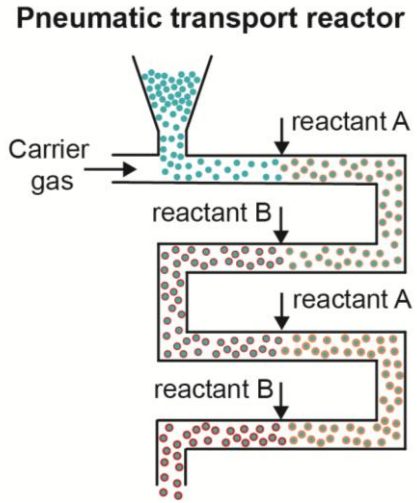
- ≥ 30% reduction in Ir (< 0.5 mg/cm² via ALD)
- ≥ 50% reduction of Pt usage by ALD Pt or no-CRM conductive, corrosion resistance coatings

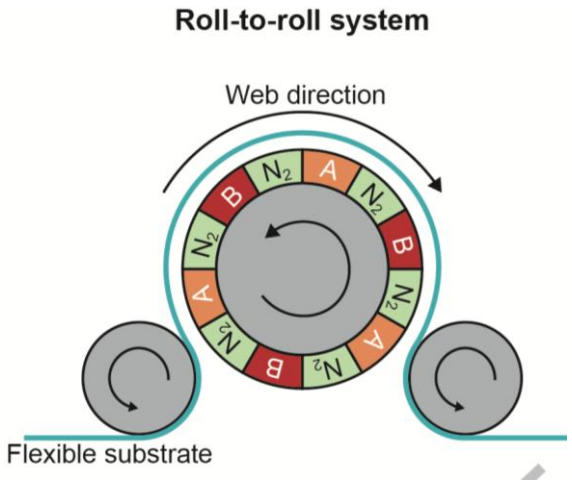
From ALD in the lab to high throughput ALD

ASM, Levitech
TU Delft
Powall
Kalpana
TU Eindhoven
TNO/Holst Centre
SparkNano
SALD
UTwente



From ALD in the lab to high throughput ALD





Key challenges towards ALD integration in a manufacturing line:

- ALD precursor volatility: for a roll-to-roll ALD system with a web width of 1 m processed at a line speed of 100 m/min, precursor consumption rate of approximately 3 kg/h
  Economical viability ALD: precursor cost and consumption (vs. surface area to be coated)
- Business case: which interface challenge can ALD address, which cannot be properly solved by other synthesis methods?

Final thoughts

The energy transition goes beyond discovering better materials.
It is also about understanding how materials behave in devices, how devices perform in systems, and whether those systems can be manufactured, deployed and sustained at scale.

Atomic-level interface engineering by ALD is only one part of that challenge.
Lessons learnt:

- A high-quality ALD layer does not necessarily lead to an efficient interface.
- Interface engineering requires balancing competing layer properties and ALD processing must be designed around device requirements & dynamic behavior of the ALD layer in (often) chemically aggressive environments.
- Next-generation energy technologies challenge ALD to move beyond atomic-scale deposition control towards device-aware interface engineering.

Acknowledgments





Kousumi Mukherjee



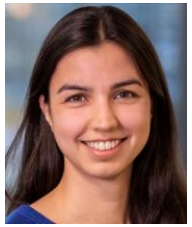
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Jan Franse Brakenhoff & Moniek Tromp (RUG, Li-ion batteries)
Mihalis Tsampas (DIFFER, water electrolysis)