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Review and challenges of aging modeling techniques for solid-state batteries

Md Sazzad Hosen

MOBI Battery Research Group, Vrije Universiteit Brussel, Pleinlaan 2, 1050 Brussel, Belgium

ABSTRACT: The state-of-the-art Gen 3 Li-ion battery degradation modeling methodologies span from electrochemical to black box approach. The methodological selection is a trade-off between computational effort, accuracy, physical insight, and data requirement. The comparatively recent Gen 4 solid-state battery aging requires advanced modeling techniques that are well-established and provide a reliable and accurate prediction.

MAIN SSB AGING MECHANISMS

- Outward diffusion of the free volume -
  - Polymer mobility: $\sigma \searrow \Longrightarrow R_{int} \nearrow$
  - Polymer matrix contraction: Effective contact area, $A_{cont}^{eff} \searrow \Longrightarrow R_{int} \nearrow$
- Cyclic deformation -
  - $A_{cont}^{eff} \searrow \Longrightarrow R_{int} \nearrow$
  - Electrolyte cracks
- Irreversible Li intercalation -
  - Loss of active material
- Dendrites growth
- Electrode particles pulverization
  - Vacant cavities $\Longrightarrow A_{cont}^{eff} \searrow \Longrightarrow R_{int} \nearrow$

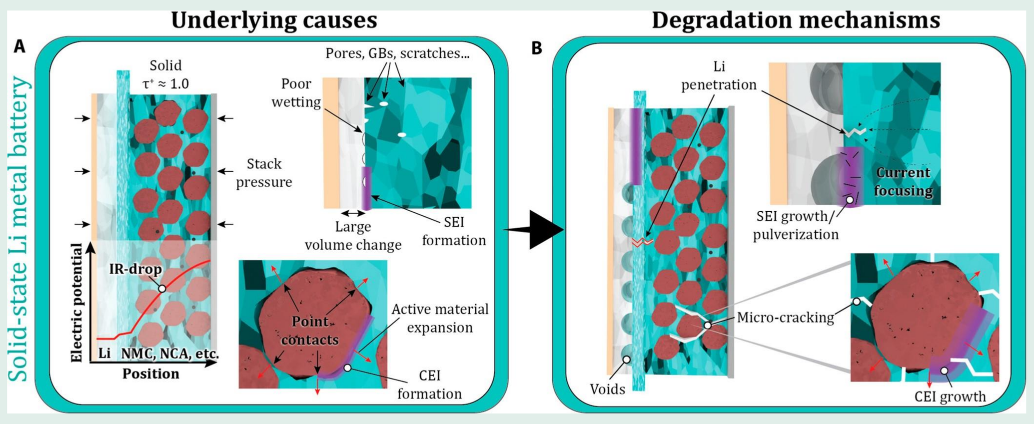

Fig 1 – Overview of some ageing mechanisms of solid-state batteries [1]

IDENTIFICATION OF A MECHANISM

Aging of solid polymer electrolyte[2]

- Poly(ethylene glycol) methacrylate with Al-PEG/B-PEG plasticizer & LiFTSI
- LFP/SPE/Li cells

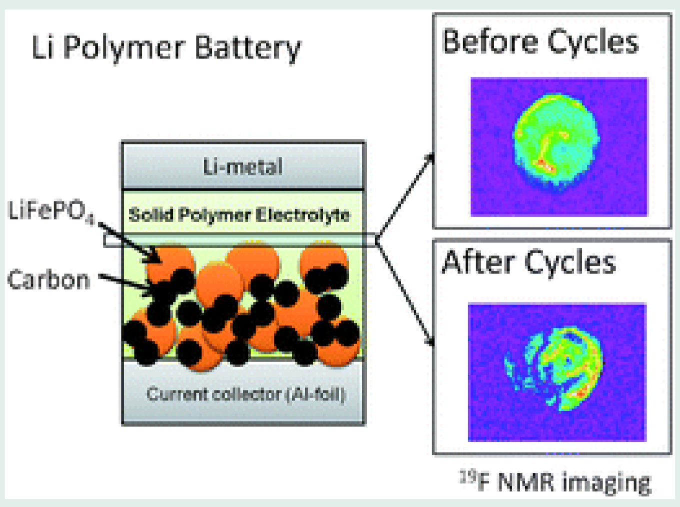

Fig 2 – NMR image of degraded SPE

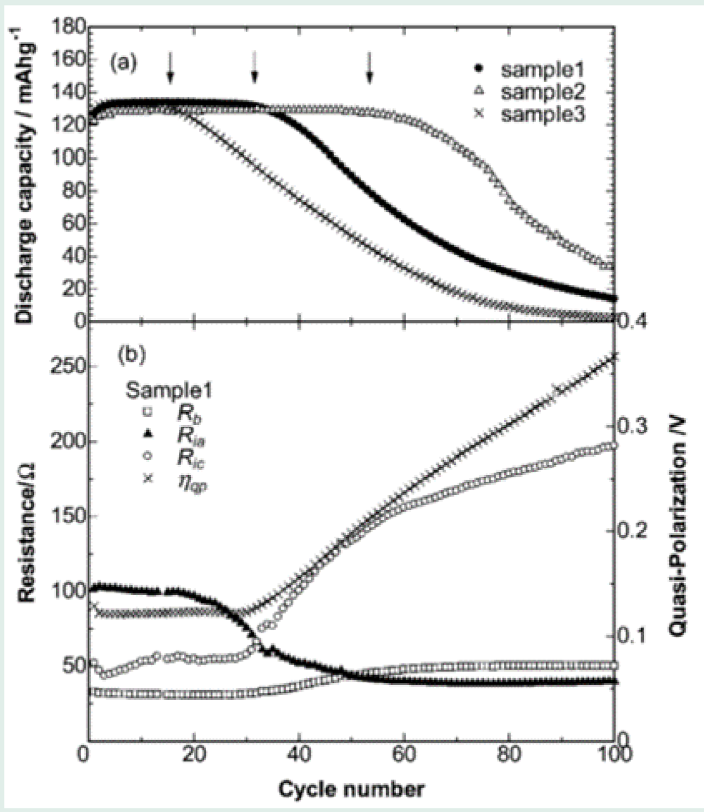

Fig 3 – (a) Cyclic performance of three identical cells (1C, 60°C); (b) Variation of internal resistances with cycling

- ✓ Pulverization of cathodic particles
  - Decrease in thickness
  - Vacant cavities: local issue
- ✓ Capacity fading and internal resistance rising related to cavities (loss of contact) and to the local decomposition of the anions due to uneven electric flow (caused by cavities)

RUL PREDICTION

A data-driven approach for aging model development[3]

- LFP/Li & NMC/Li cells
- Cross-linked nanocomposite polymer electrolyte: PPO and PEO-based block polymer with SiO_2 nanoparticles
- Machine Learning: Symbolic Regression

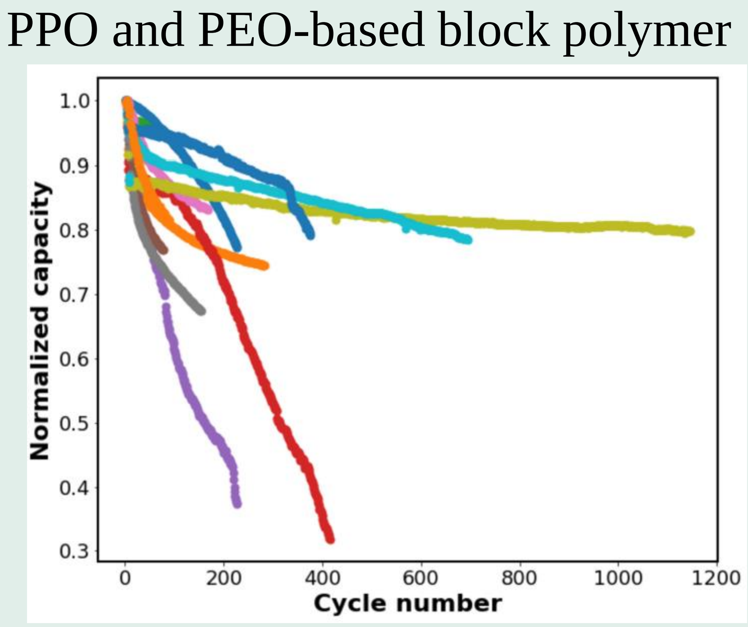

Fig 4 - Discharge capacity of cycled solid-state batteries

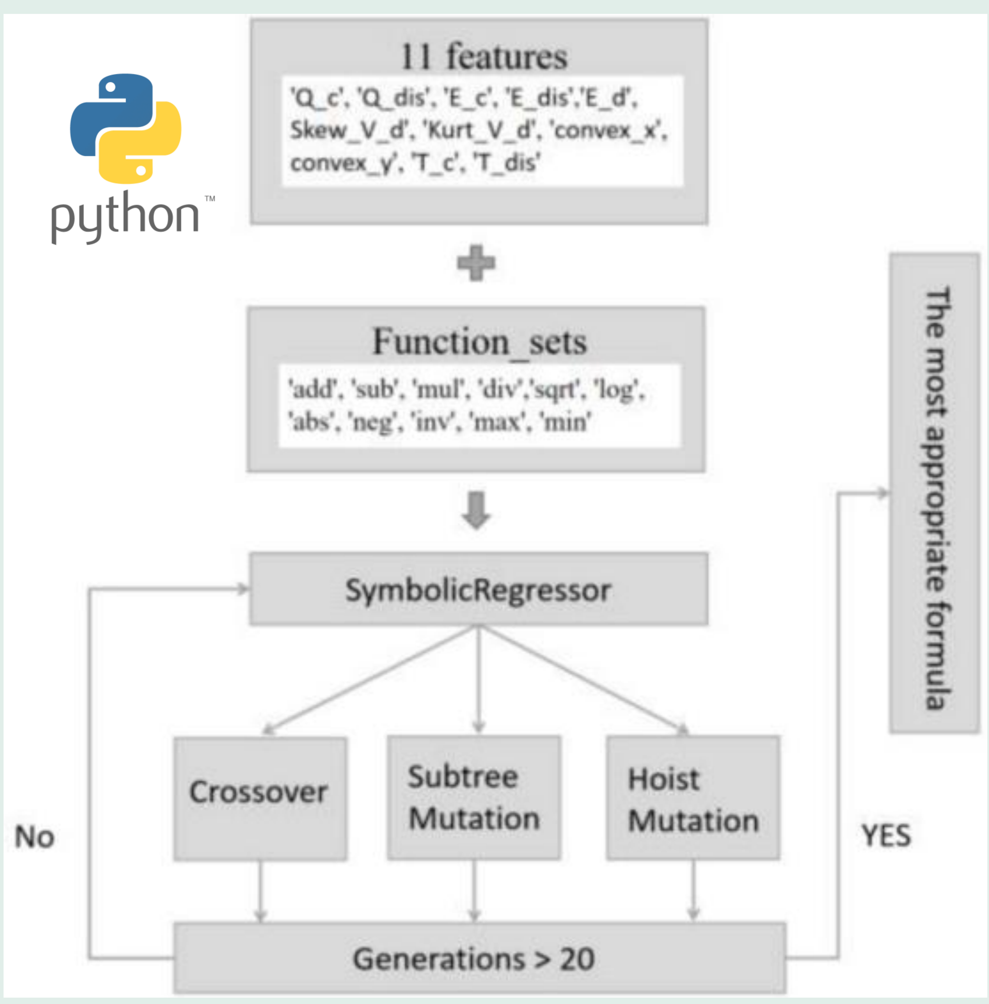

Fig 5 - Computational routine of Symbolic Regression model

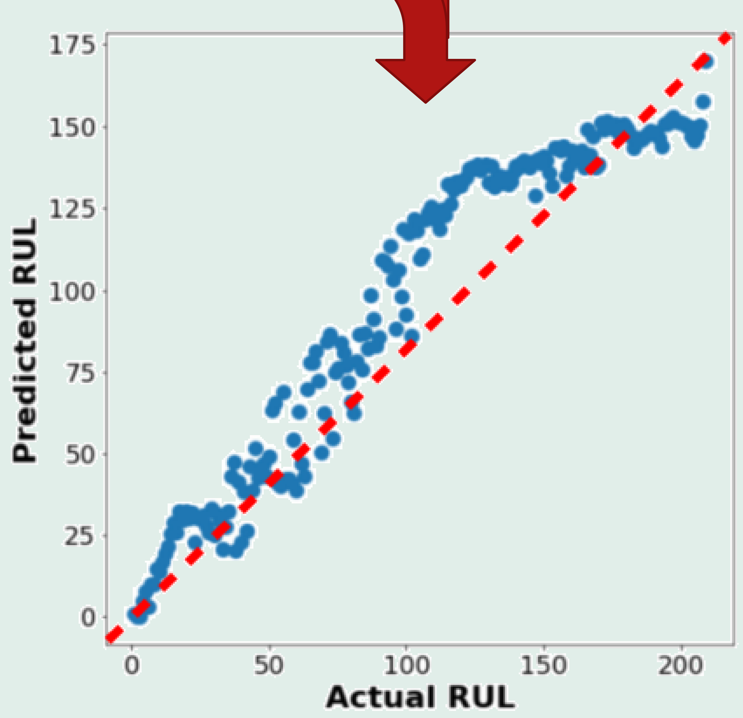

Fig 4 – Comparison of predicted and actual RUL

- ✓ RUL of different battery types predicted
- ✓ $R^2 = 0.91$
- ✓ Better prediction performance with respect to other ML methods: GPR, EN and SVR.

CAPACITY FADE PREDICTION

Continuum modeling[4]

- LCO/Li with LiTFSI + poly(AN-co-BuA) electrolyte
- Three homogeneous domains → 1D model
- $A/A_0 \propto V/V_0 \propto$ free volume
- Relation between free volume and discharging capacity
  - Model of SPE aging

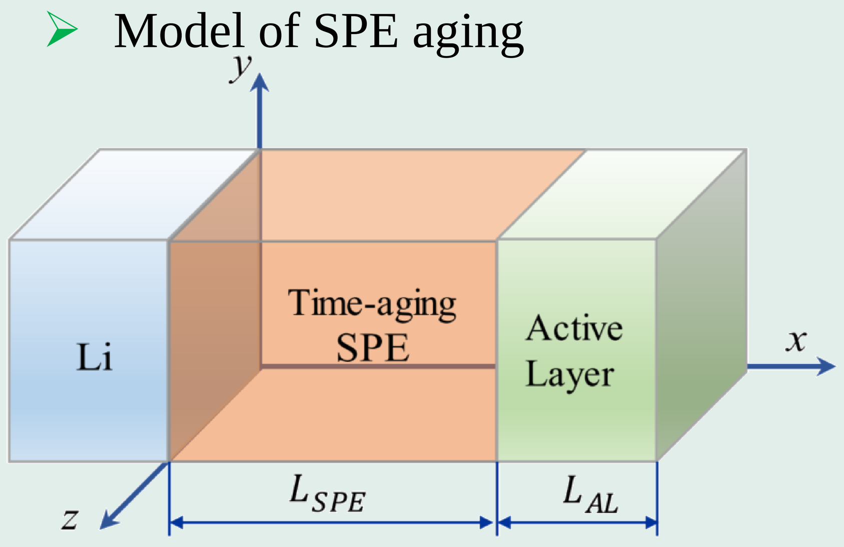

Fig 6 – 3D scheme of a solid-state battery with a solid polymer electrolyte

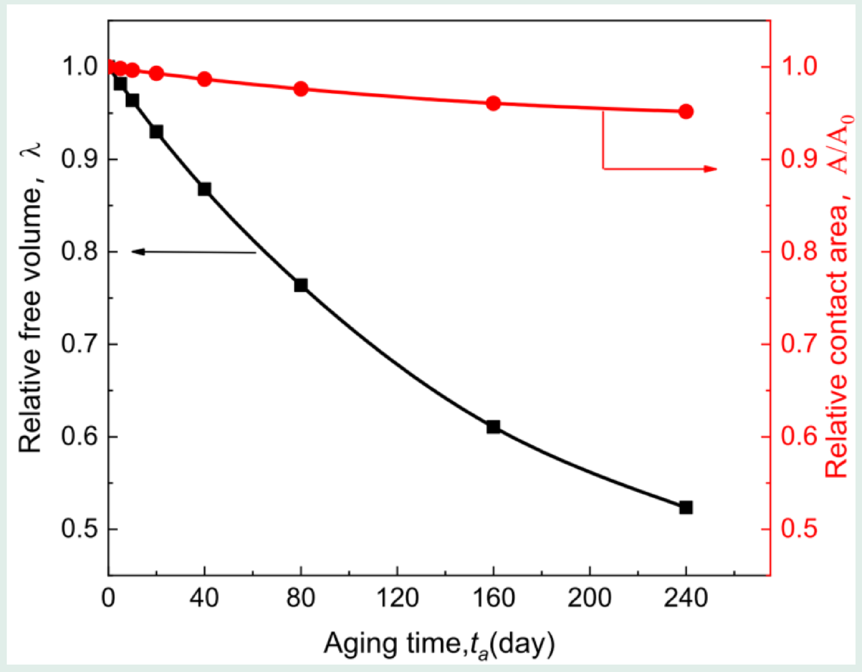

Fig 7 – Effect of modelled polymer relaxation on its free volume and contact area with electrode

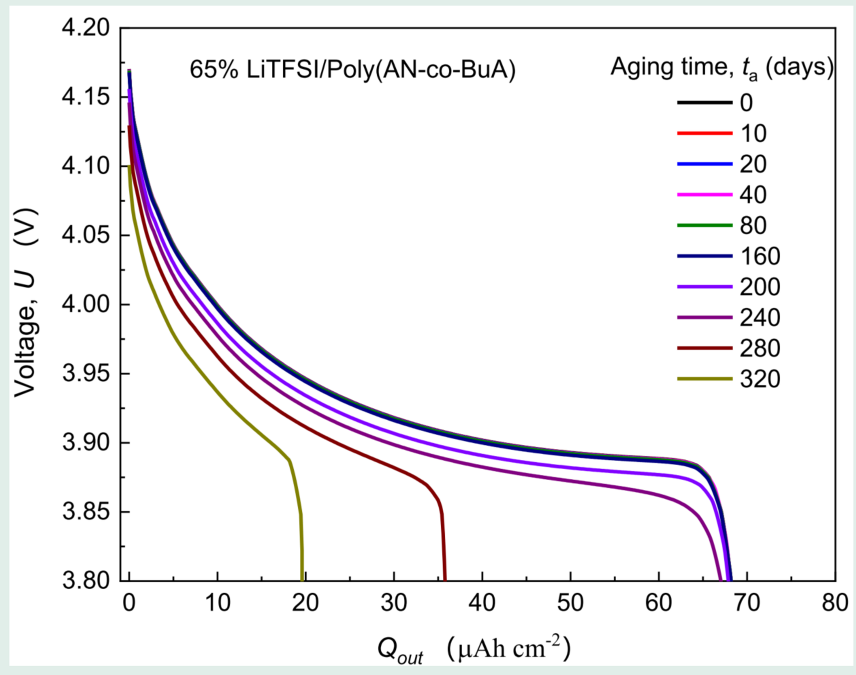

Fig 8 – Effect of time-ageing on discharge curve at 1C rate

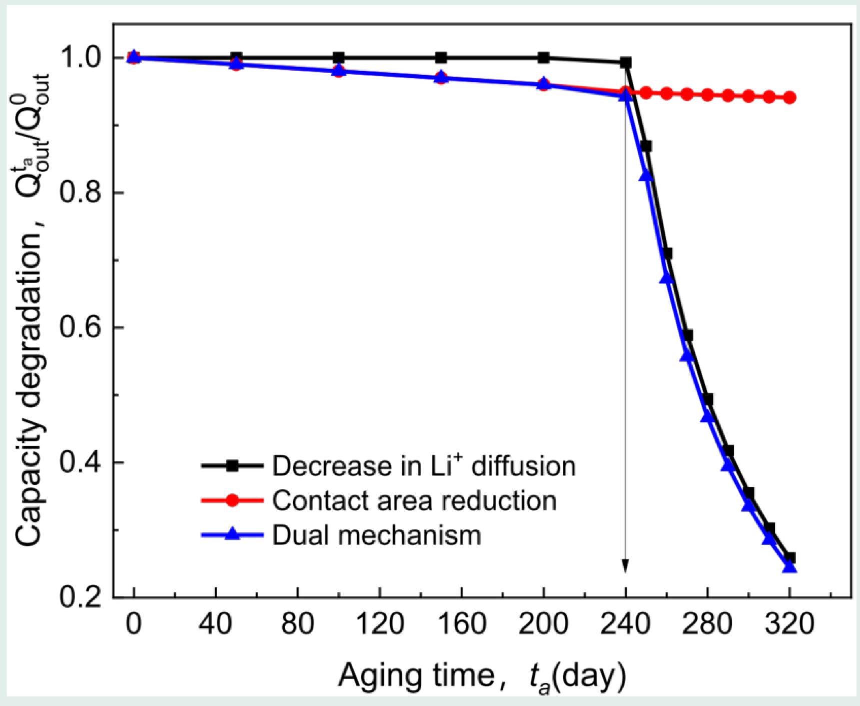

Fig 9 – Effect of electrolyte time-aging on discharge capacity

- ✓ Time-domain aging model
- ✓ Impact of free volume decrease
  - Loss of ionic conductivity
  - Loss of effective contact area
- ✓ Influence of other factors (T, C-rate, salt concentration, etc.)

TAKEAWAYS

- Model-based approach: high interpretability, less complexity
- Data-driven approach: high accuracy, less interpretability
- Most of the existing literature focus on understanding aging mechanisms and improving the ionic conductivity
  - Very few studies on modeling SSB aging
- Hybrid Solid Electrolyte (HSE): polymer matrix & inorganic filler
  - Combines → the flexibility of polymer and advantages of inorganic electrolyte
  - Promising but not mature → aging not still modeled
- Open-access dataset is rarely available for Gen 4 batteries.

FUTURE WORK & ACKNOWLEDGEMENT

- Prototyping of monolayer pouch cells of NMC811 cathode and HSE (inorganic and polymer) for an experimental study.
- Develop cycling and performance data of SSB prototypes (anode-less) that are reproducible and of high quality.
- Demonstrate model-based aging prediction with high accuracy within the scope of the **AM4BAT** project framework.
- This project has received funding from the European Union's Horizon Europe research and innovation program under grant agreement No 101069756.

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VUB — mobi ELECTROMOBILITY RESEARCH CENTRE

Md Sazzad Hosen
Senior Researcher
Md.Sazzad.Hosen@vub.be

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