
DTU

High-throughput workflows with ASE and Fireworks

Felix Tim Bölle

An ASE-journey towards automating migration barrier calculations for battery materials

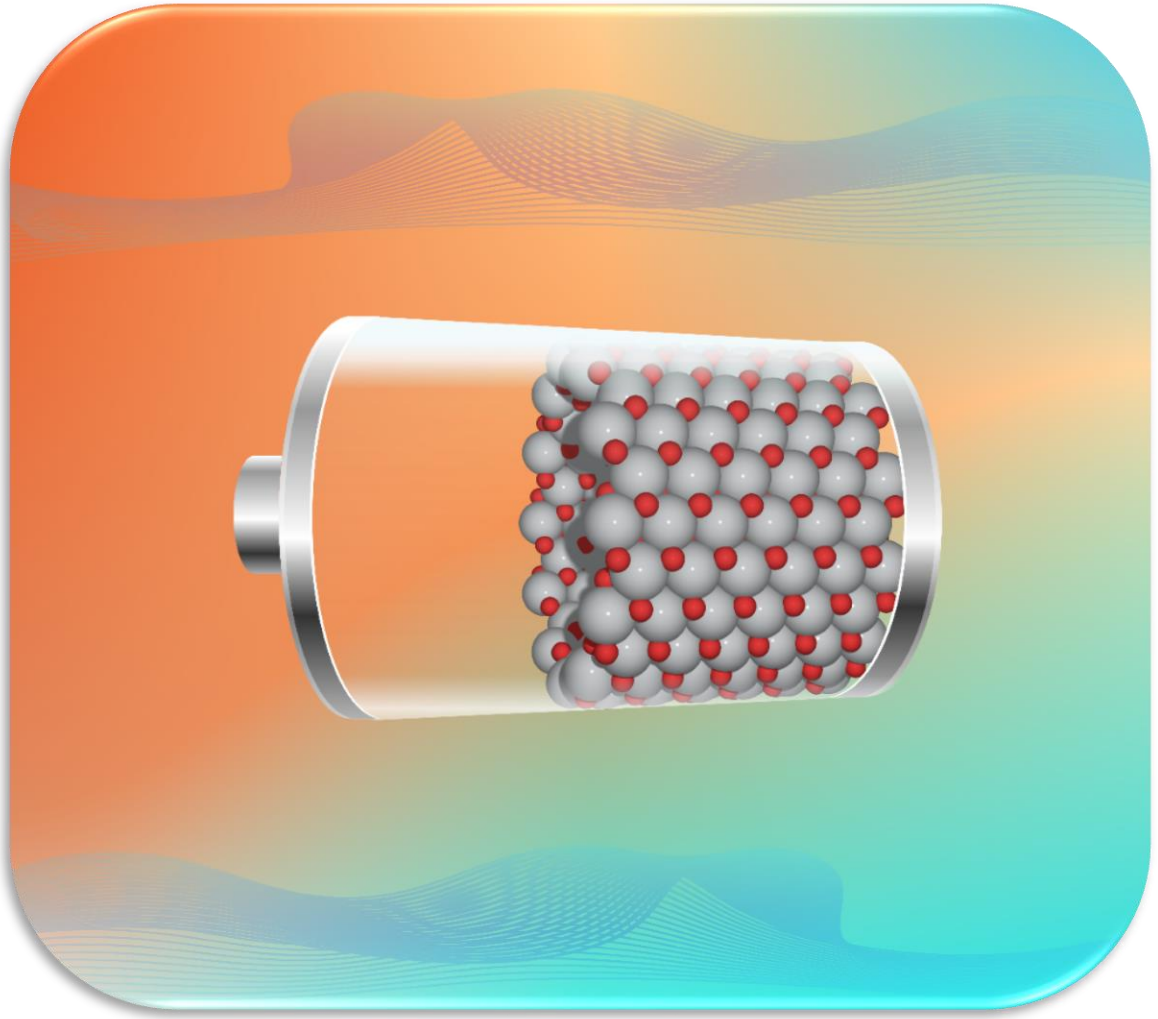
Outline

I. **Automating migration barriers**

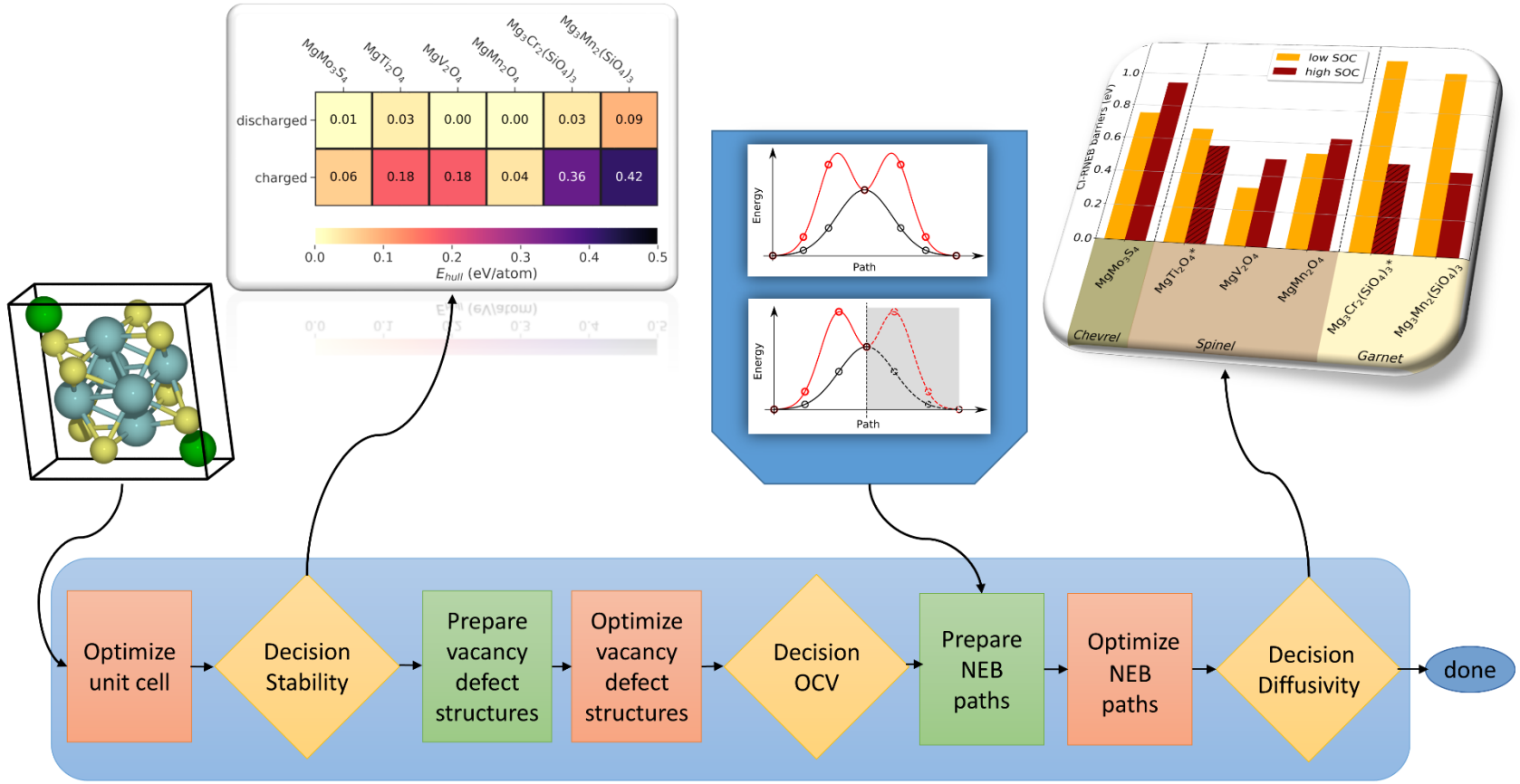
II. **Methods**
- How to choose the right tool?
- Limits of the chosen tools

III. **Workflow for calculating ion insertion battery properties**
- Results
- Automation, then what?

IV. **Lessons learned**
- The tool I am still looking for



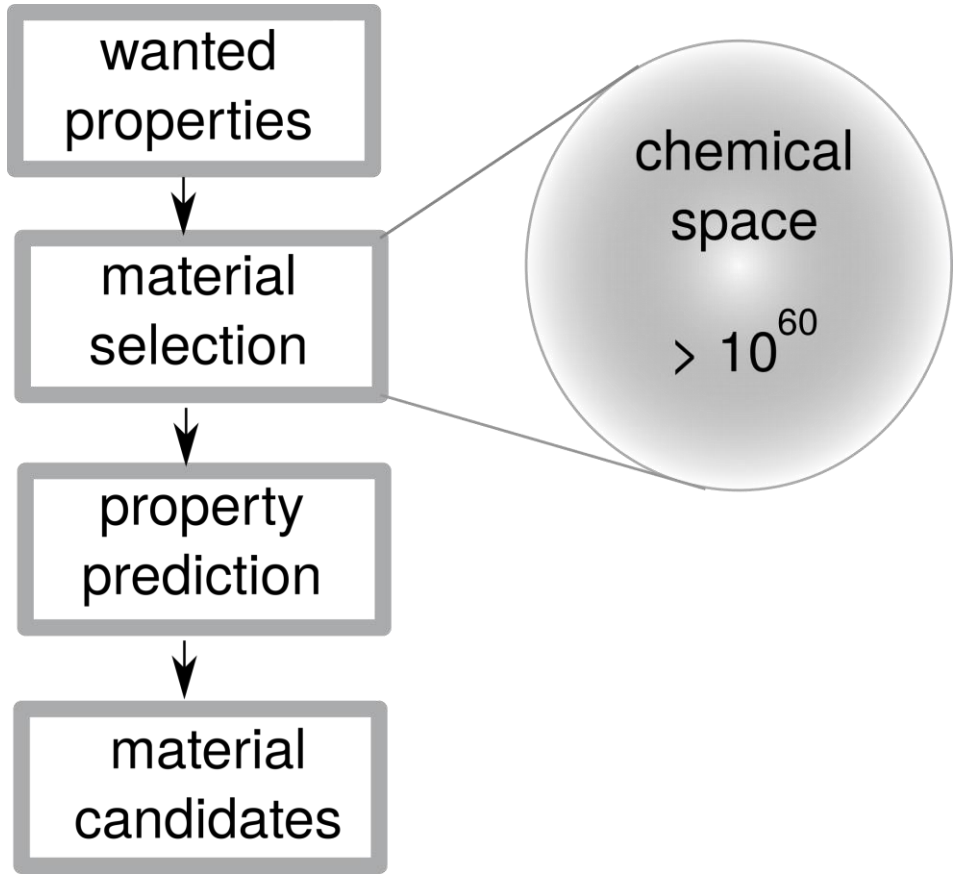
Automating migration barriers



Bölle, Mathiesen, Nielsen, Vegge, García-Lastra, Castelli. *Batteries and Supercaps*.; 3(6), 470-470 (2020).

Methods – scientific workflows and DFT

- Simulations performed using Density Functional Theory (DFT)
  - Solving the electronic structure of a system starting from first-principles
- Problems for accelerated discovery
  - Access to data
  - Standards for data
- Scientific workflows for reproducible data
  - Utilize datasets
  - Generate new data to extend dataset



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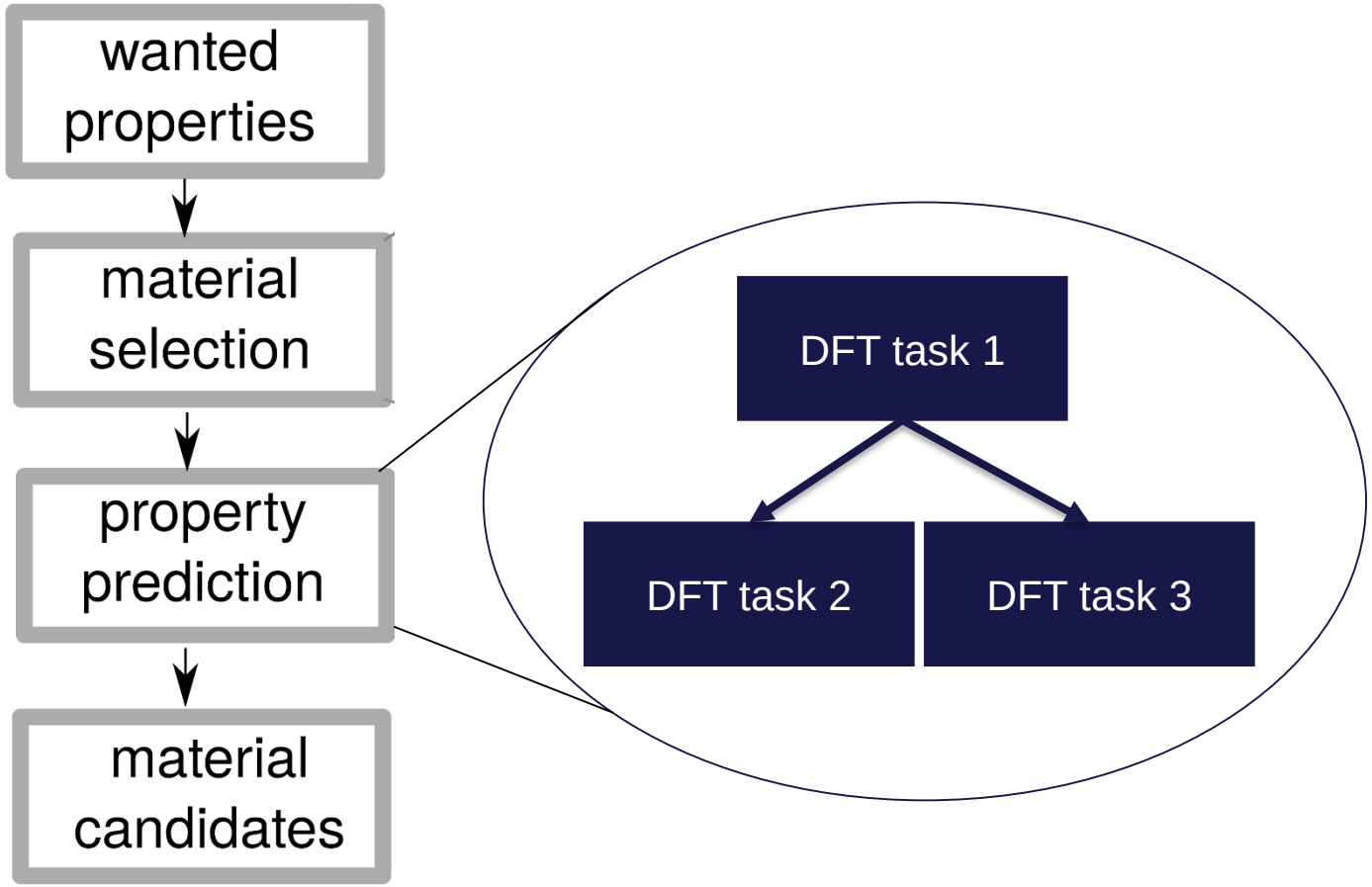

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- Reproducible data
  - Same workflow input leads to same workflow output
  - No manual user intervention
- What is called reproducible?

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$ workflow run user_input.json
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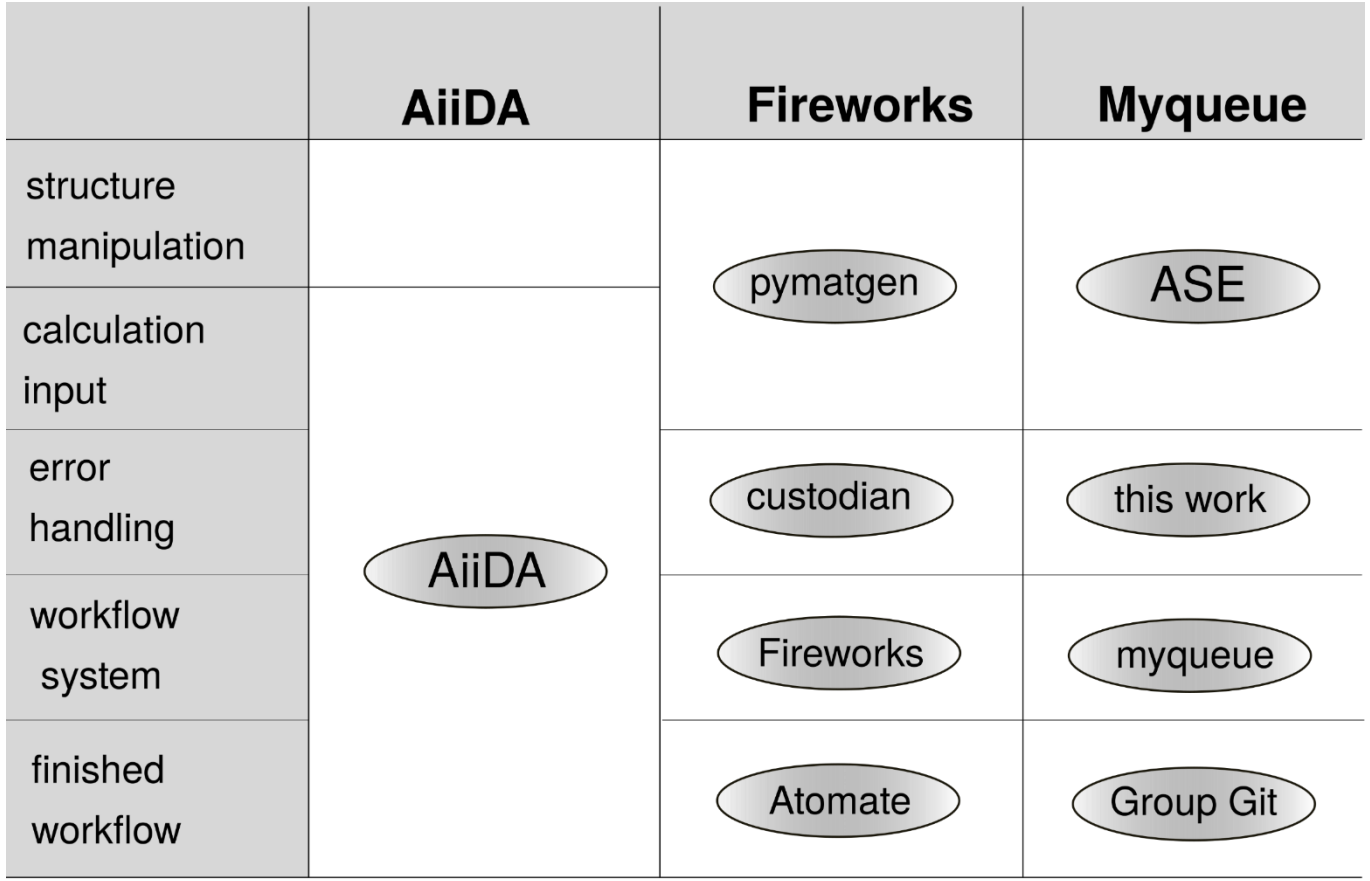
- Helpful software tools exist/are being developed to support researchers
- Common “workflow-language”



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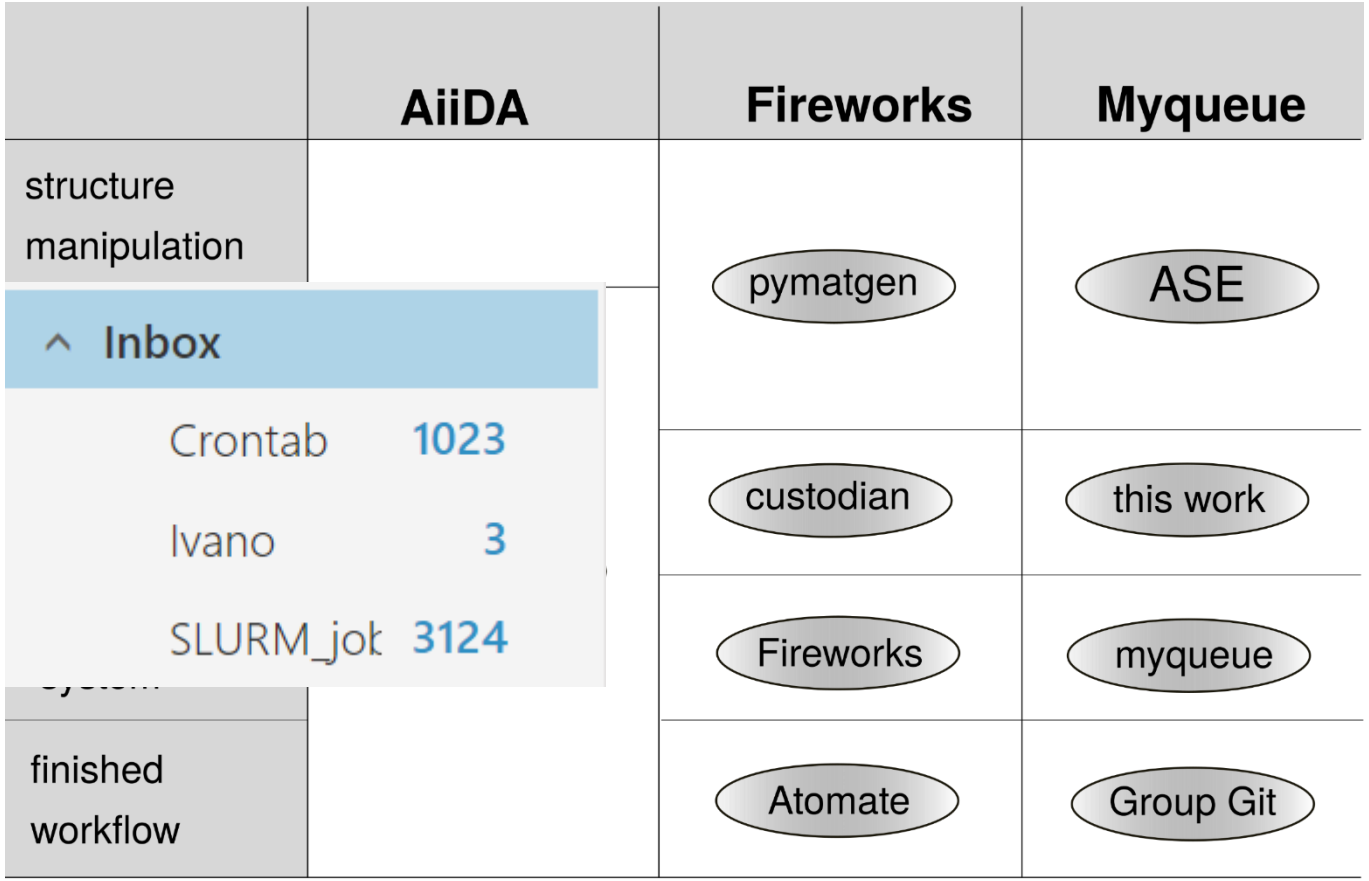

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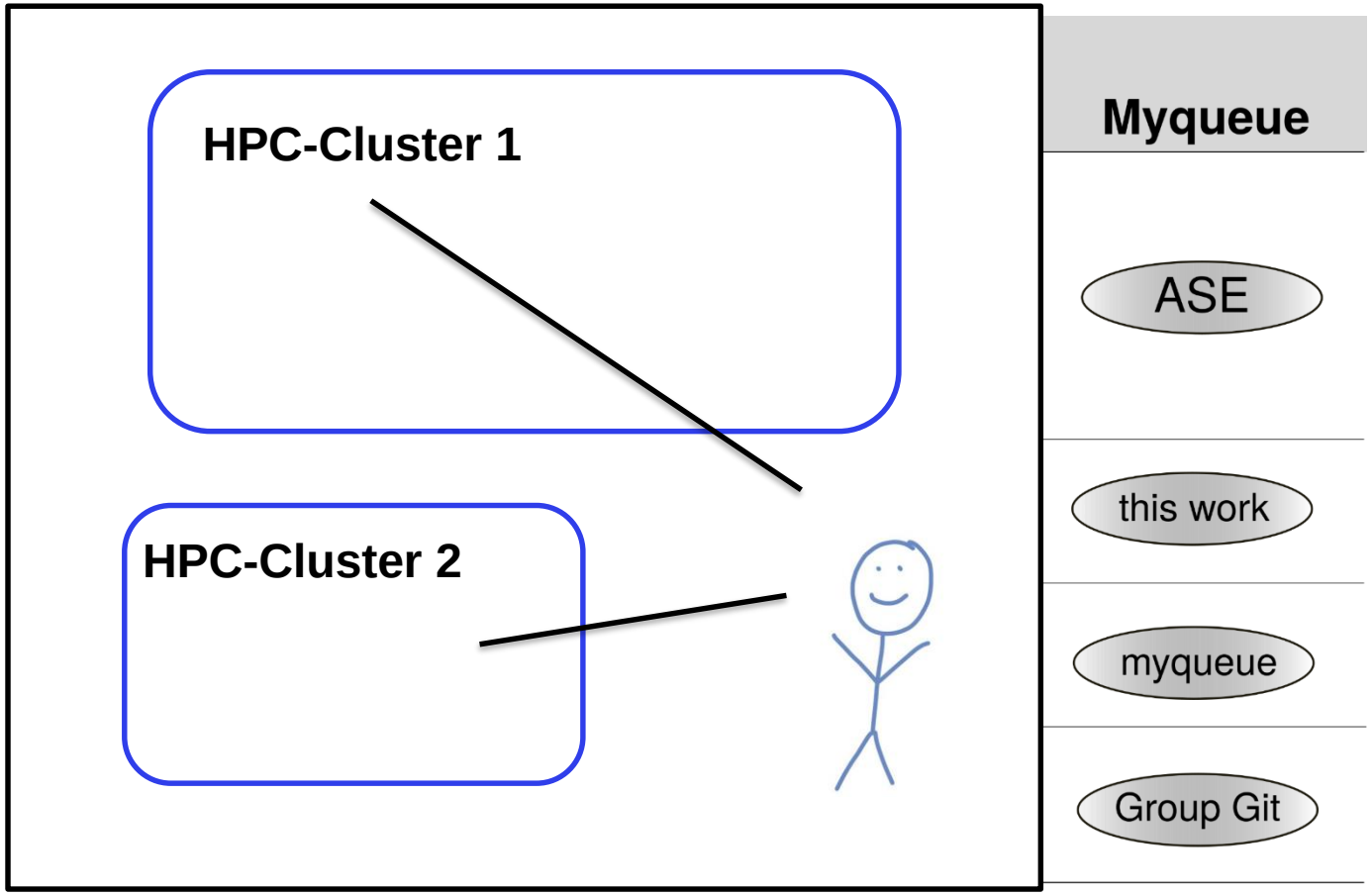
- What are colleagues doing?
- What am I missing?



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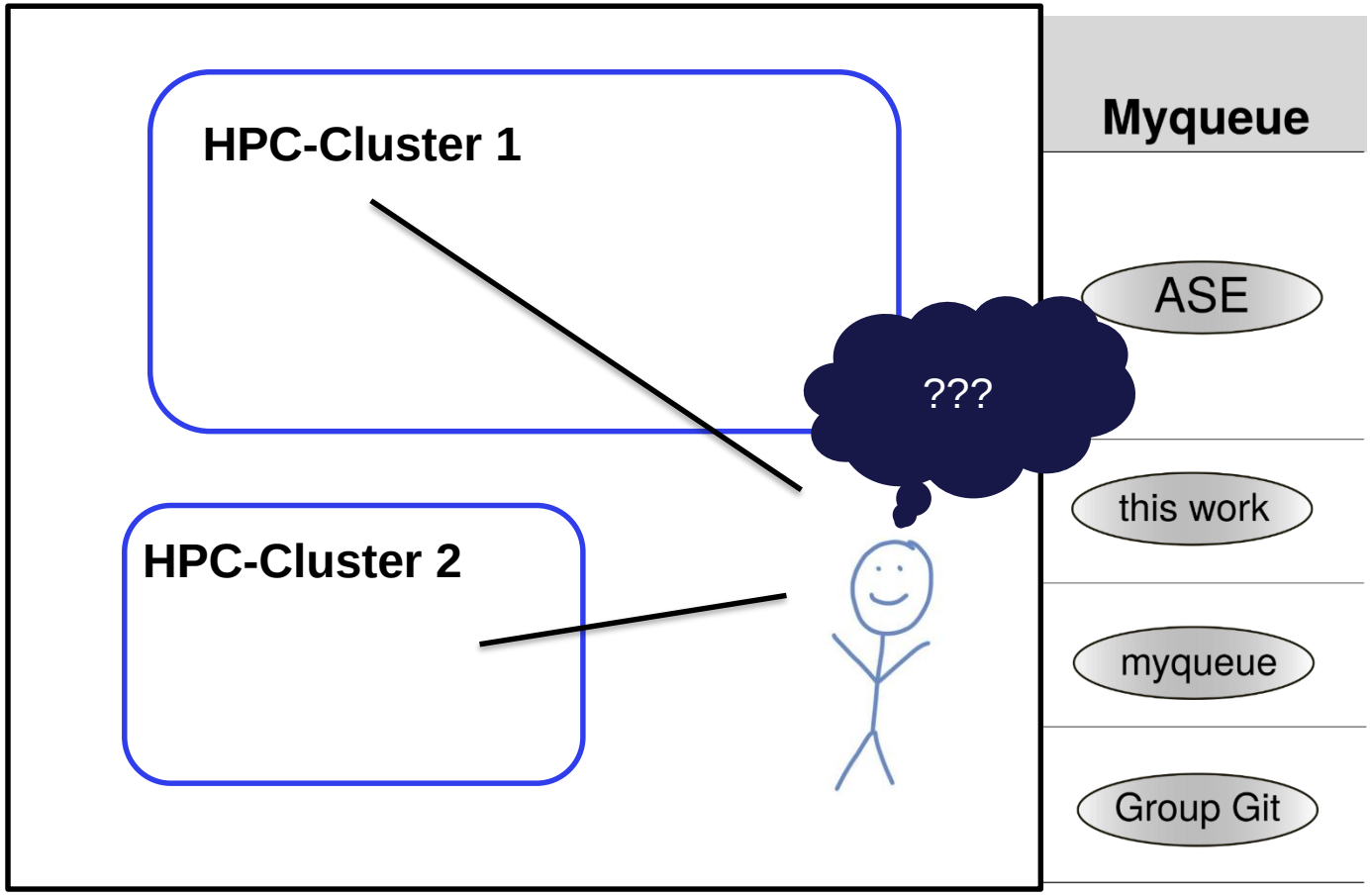

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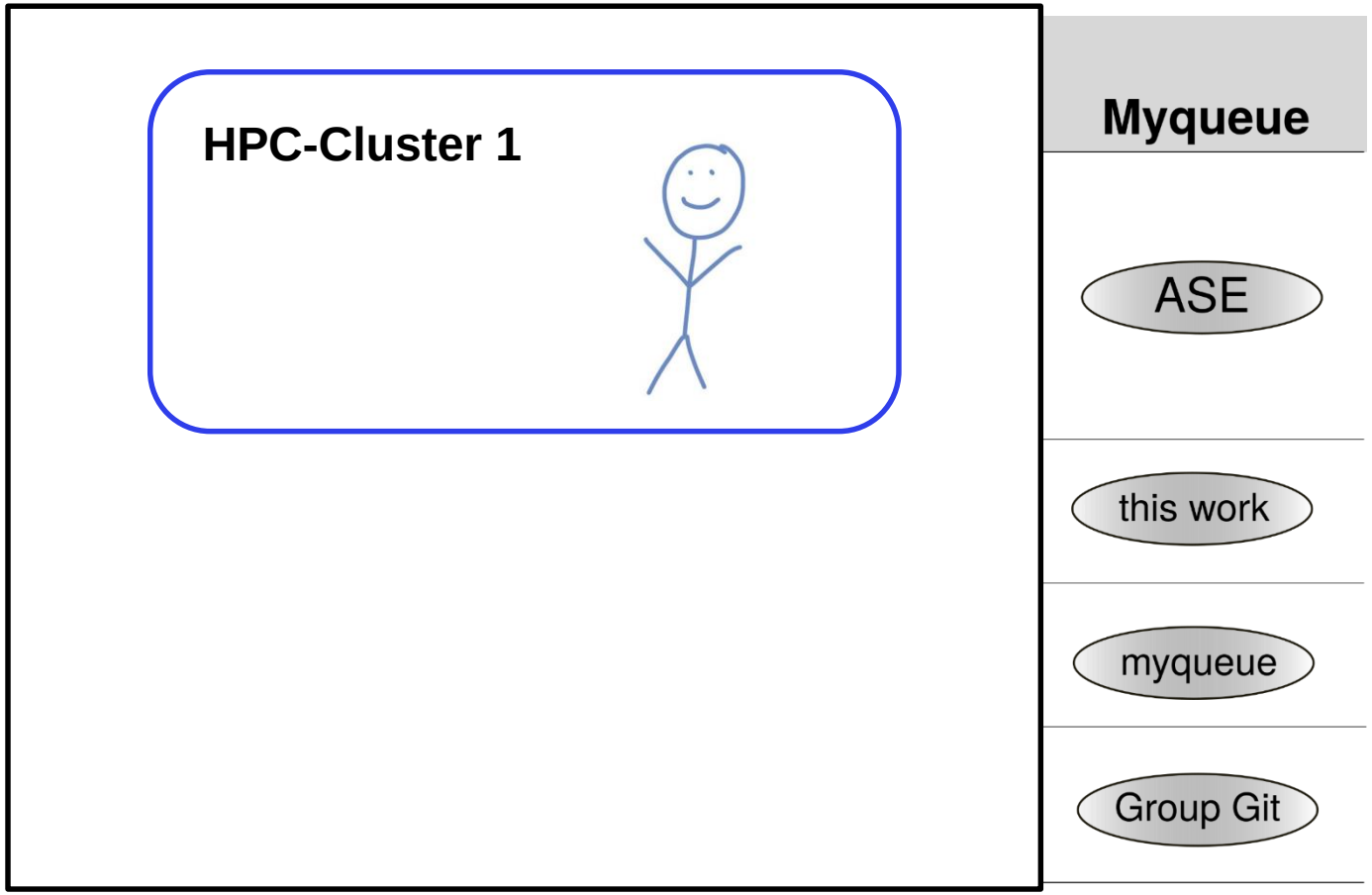

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Methods – scientific workflow tools

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  - Missing piece for fully automatic workflows
- Inspired by CUSTODIAN and implemented for ASE
  - Did not exist for ASE
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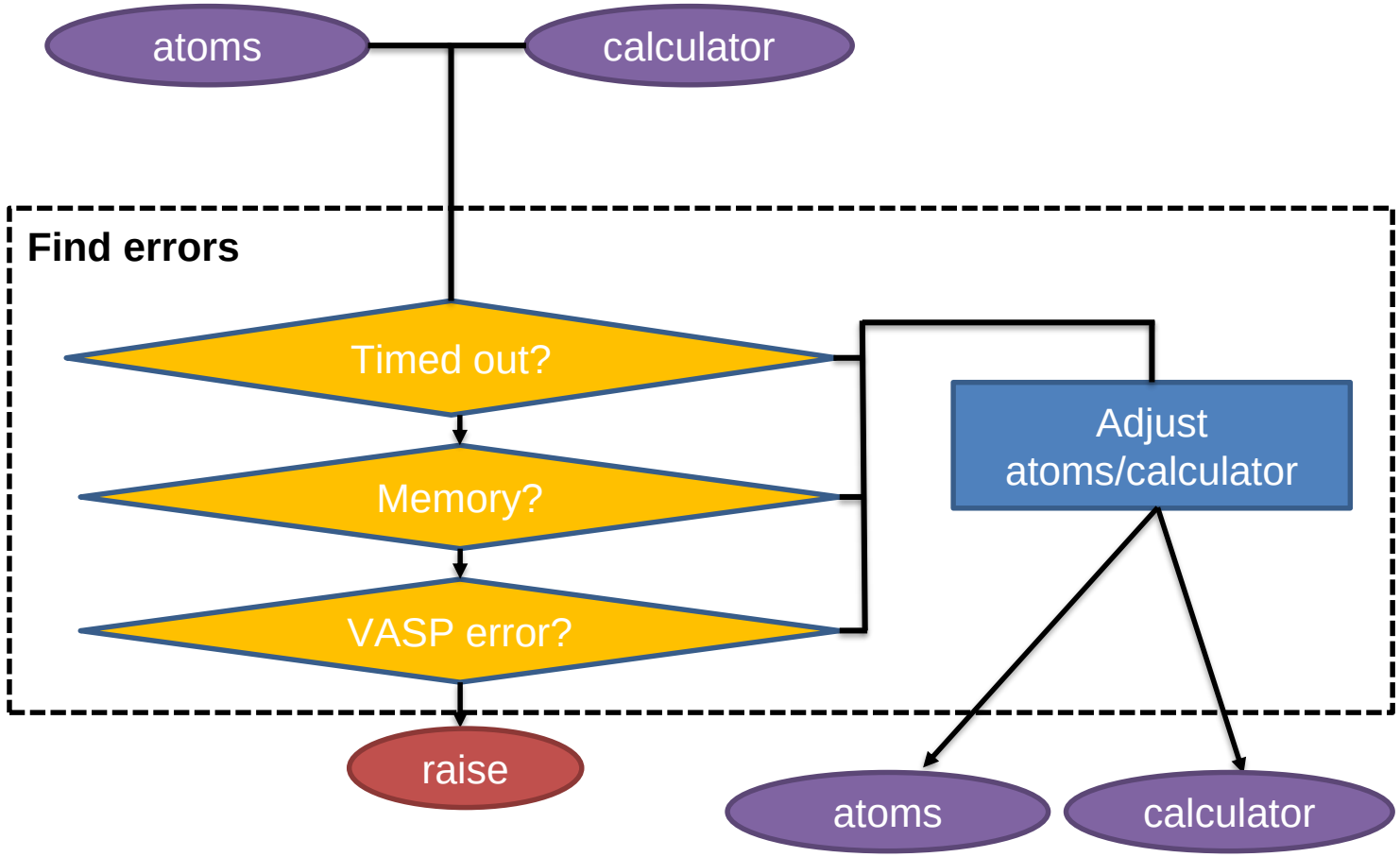

[1] Bölle et al. Batteries & Supercaps 3(6), 488-498 (2020)

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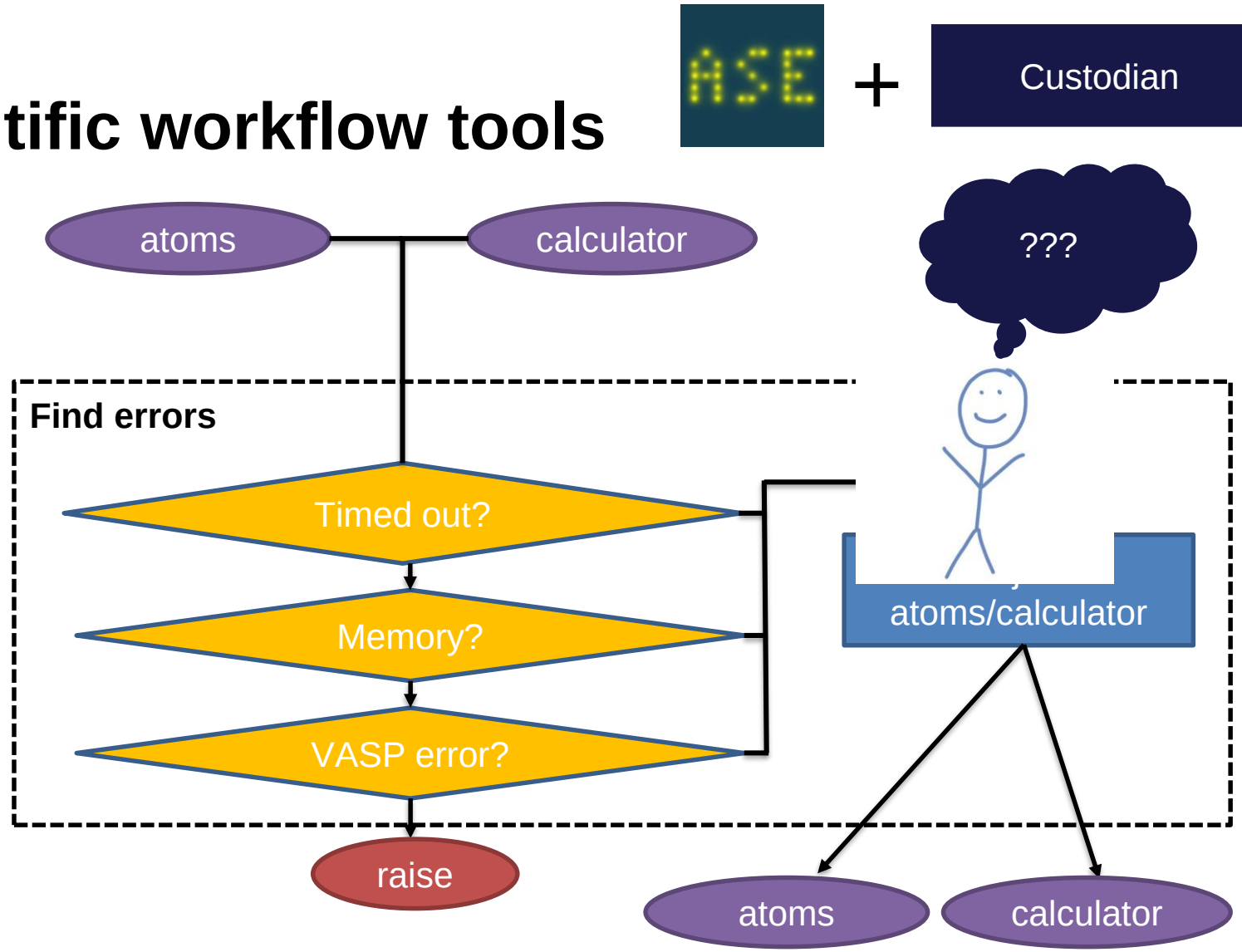

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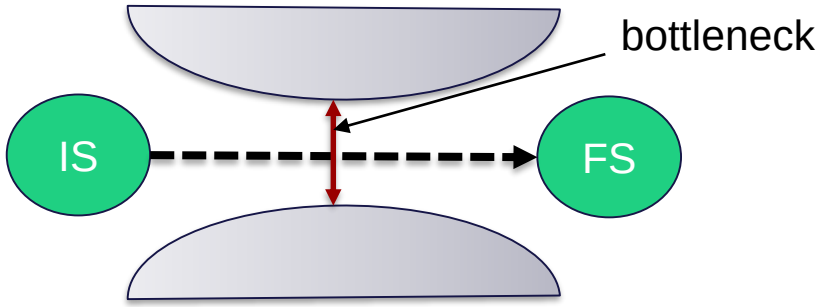
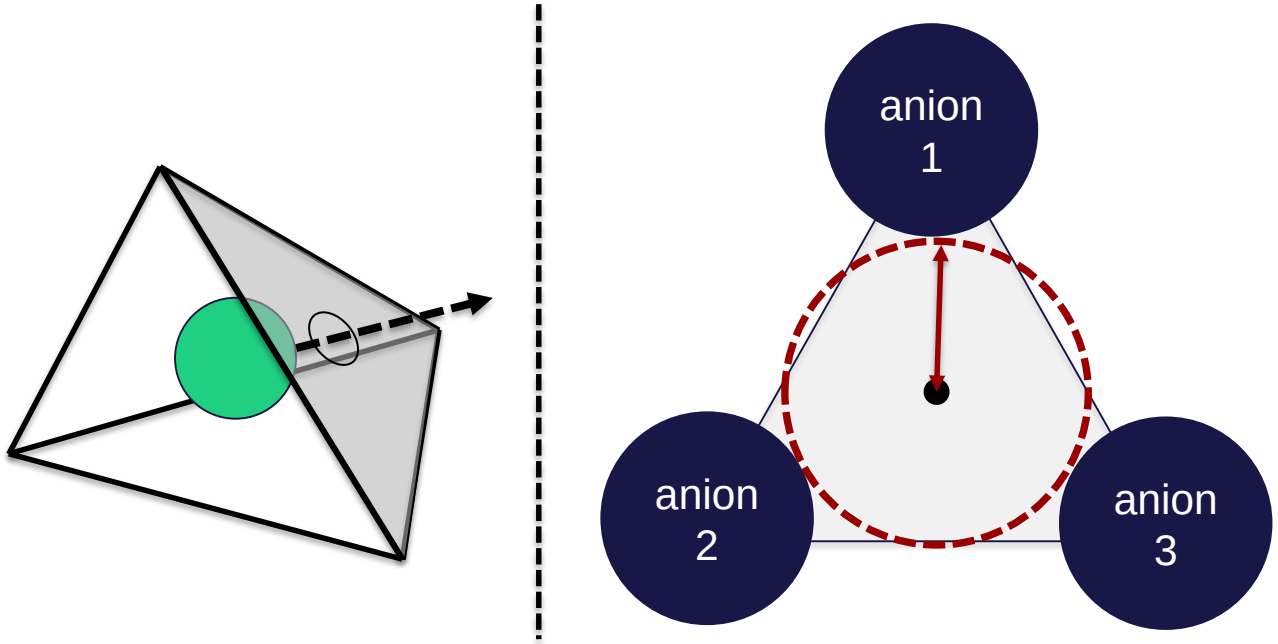
Insertion batteries – Results



- NEB calculations 3-4 orders of magnitude slower than unit cell relaxation
- Find descriptors to predict diffusion barriers?
- How to predict **transition state** before relaxing the path?
- Find the **bottleneck** of the path
  - Identify possible intermediate interstitial sites
  - Identify all neighbors
  - Calculate critical radius for each face

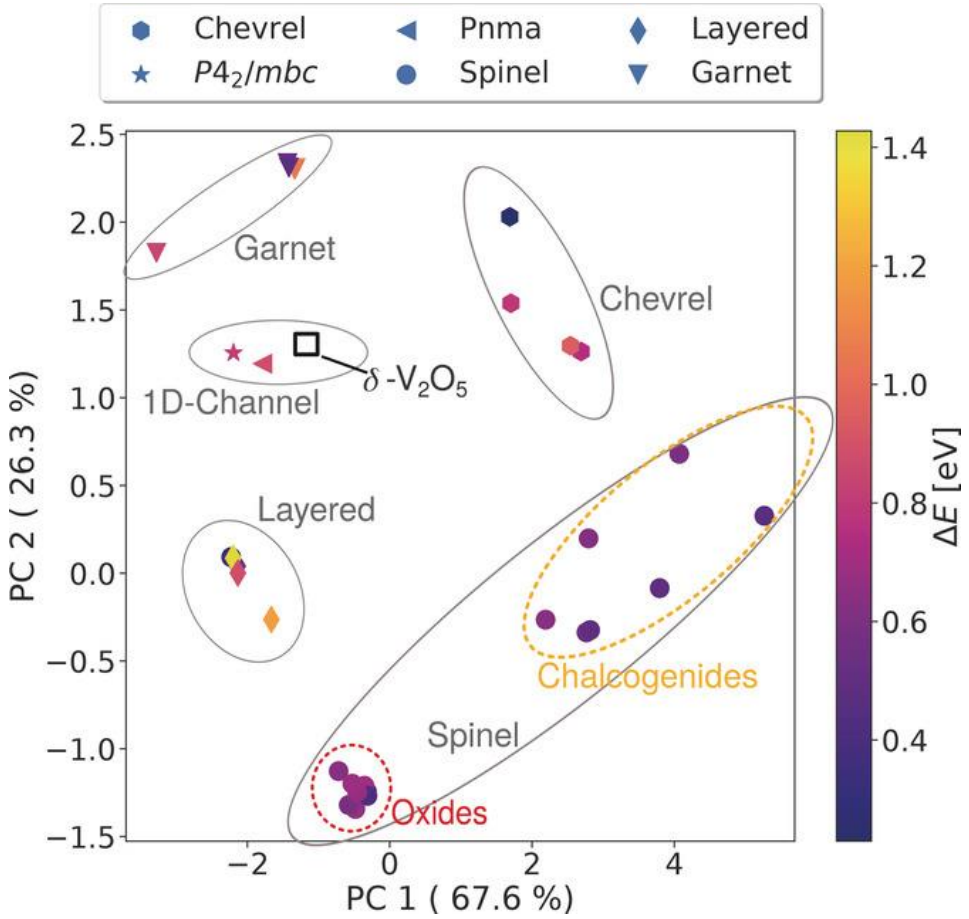


V. A. Blatov*. *Crystallography reviews*, 10(4), 249-318. (2004)

Bölle, Bhowmik , Vegge, García Lastra & Castelli. Batteries & Supercaps 4(9), 1516-1524 (2021).

Insertion batteries – results for Mg-batteries

- Correlation of features is promising
  – Nevertheless: only 40 data points
- Sub-group diffusion topologies using Principal Component Analysis
- PCA based on six geometrical features
- Make screening more efficient
  – Autonomous agents
- Automation leads to more work



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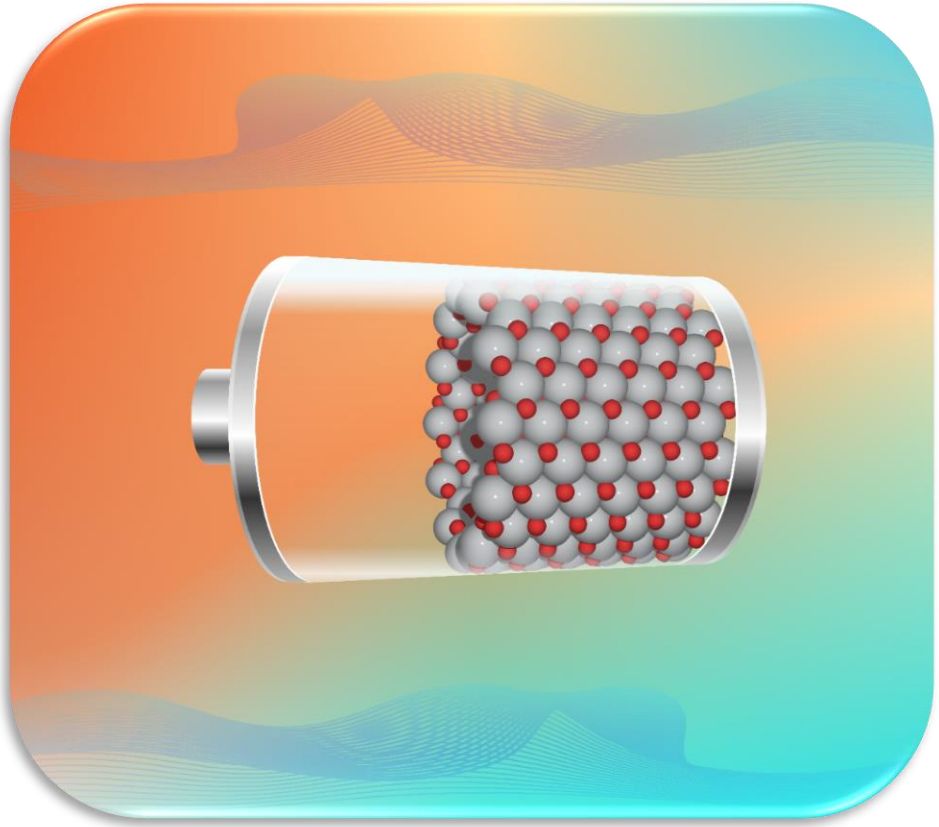
Conclusions or lessons learned

- **Choose tools already available**

- **Keep it modular** on all levels
  - Every task is a class (see ASR recipes)
  - ASE/pymatgen
    - Error handler/Custodian
      - MyQueue/Fireworks
        - » Your workflow

- **Interaction between tasks** is tricky

- **Error handling is key**
  - ASE + Custodian ?
- **Reproducible data** for Machine Learning



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Thank you!

Ivano E. Castelli

Tejs Vegge

Juan-Maria G. Lastra

Nicolai R. Mathiesen
Alexander J. Nielsen
Andrea Fedrigucci
CAMD

AiMade
Autonomous Materials Discovery

Department of Energy Conversion and Storage,
Technical University of Denmark

MetaBatt

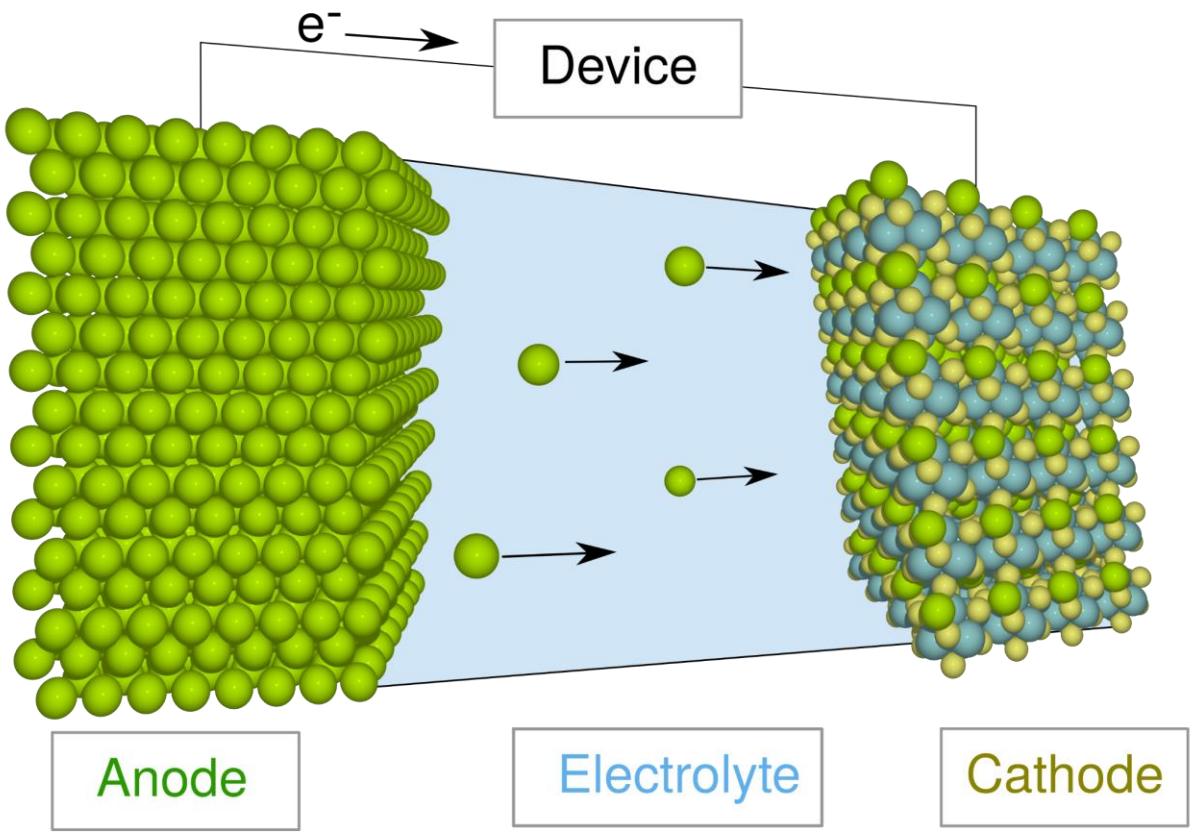
Danish ERC Programme, project: Multiscale Design of Electrochemical Metamaterials.

Paper: https://doi.org/10.1002/batt.201900152
Repository: https://gitlab.com/asc-dtu/workflows/ion-insertion-battery-workflow
Documentation: https://asc-dtu.gitlab.io/workflows/ion-insertion-battery-workflow/

APPENDIX

Insertion batteries - Introduction

- Why multivalent?
  - Increased capacities
  - Abundant resources
- Main challenge cathode?
  - Strong interaction of magnesium ion with its environment, slow diffusion
- State of the art material?
  - Chevrel phase $Mg_xMo_6S_8$ ($0 < x_{Mg} \leq 2$)
  - Open circuit voltage ~ 1.1V

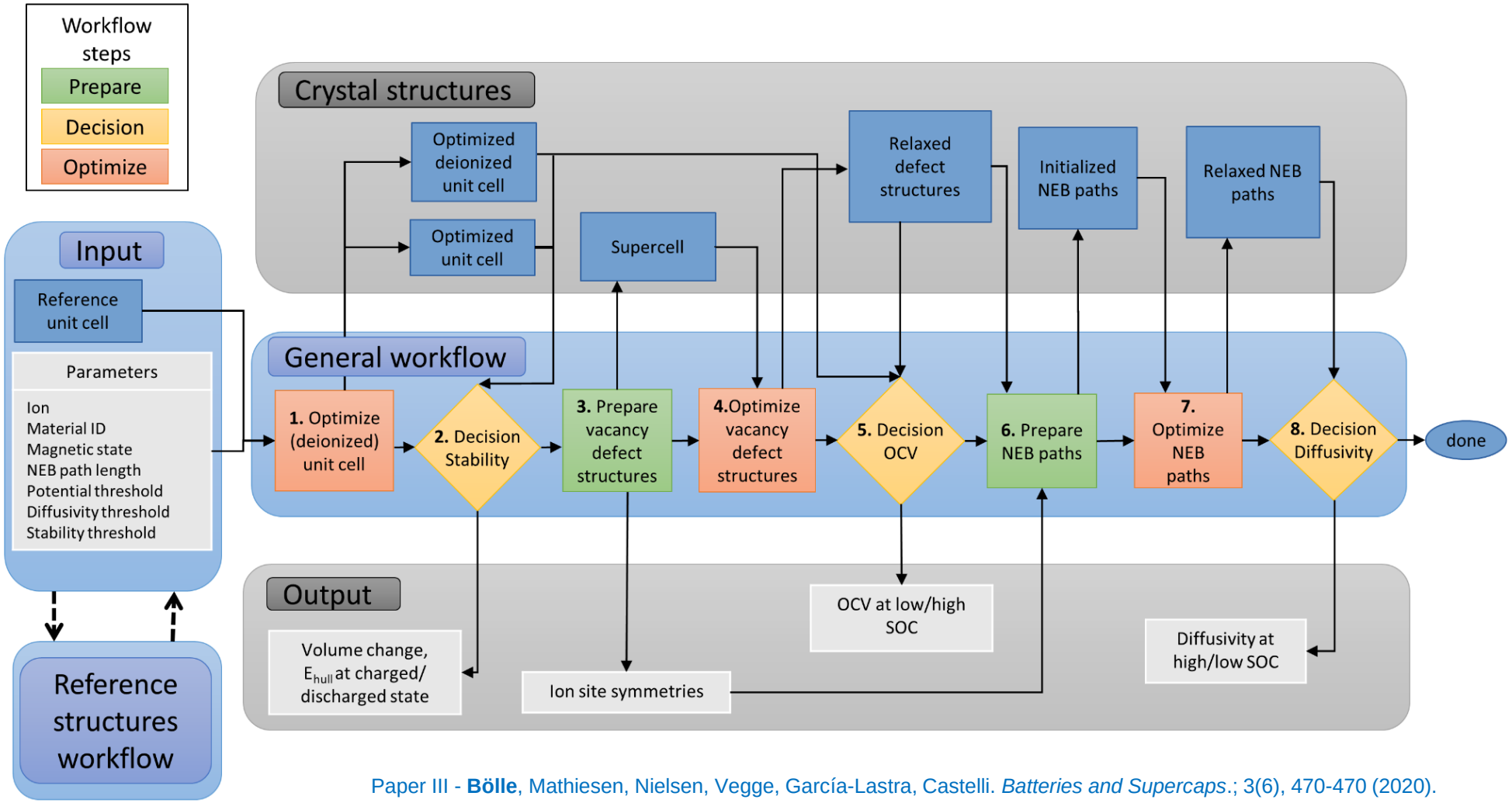


Canepa, Gautam, Hannah, Malik, Liu, Gallagher, Persson, Ceder. *Chem. Rev.*, 117, 4287–4341 (2017).

Aurbach, Schechter, Gofer, Gizbar, Turgeman, Cohen, Moshkovich, Levi. *Nature* 407, 724-727 (2000).

Insertion batteries - Introduction

- Automate calculating key properties (DFT) for magnesium ion insertion cathodes

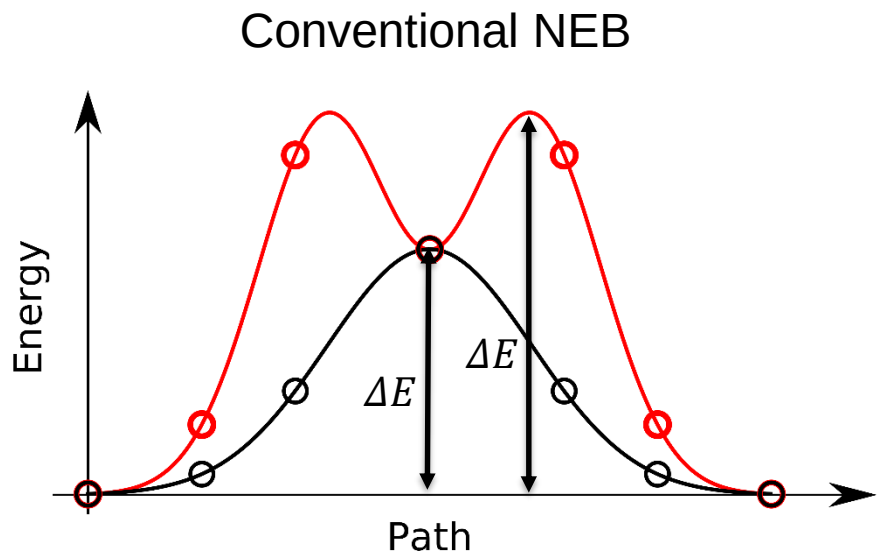


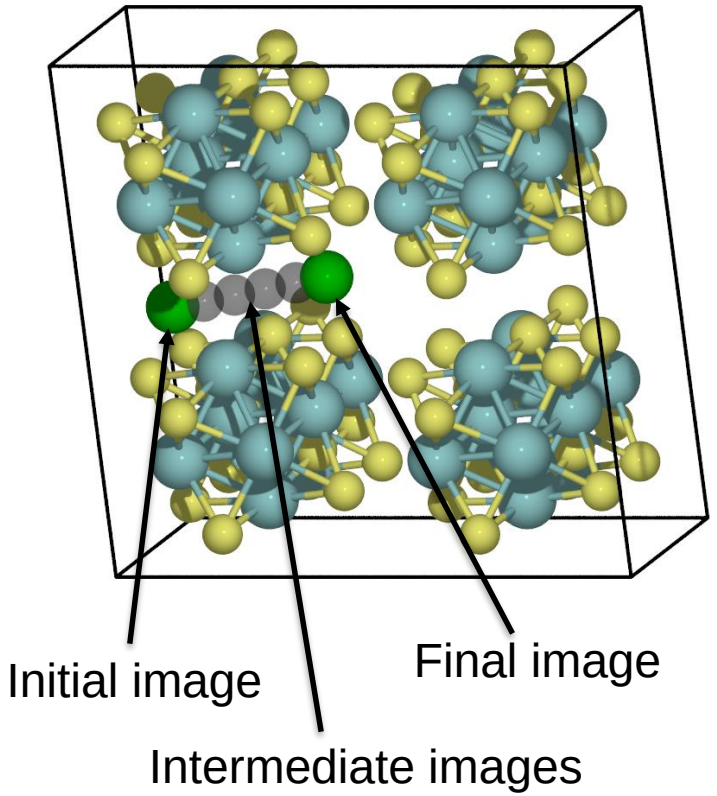
Paper III - **Bölle**, Mathiesen, Nielsen, Vegge, García-Lastra, Castelli. *Batteries and Supercaps*.; 3(6), 470-470 (2020).

Insertion batteries – NEB logic

5. Prepare NEB paths

- Running a full nudged elastic band (NEB) calculation is expensive – 3-4 orders of magnitude slower than unit cell relaxation
- Reduce the number of images to calculate using reflection symmetry
- Reflect not only positions but also forces
- NEB barrier can be related to diffusivity





Mathiesen, Jónsson, Vegge, García-Lastra. *Journal of chemical theory and computation* ,15(5) 3215-3222, (2019).

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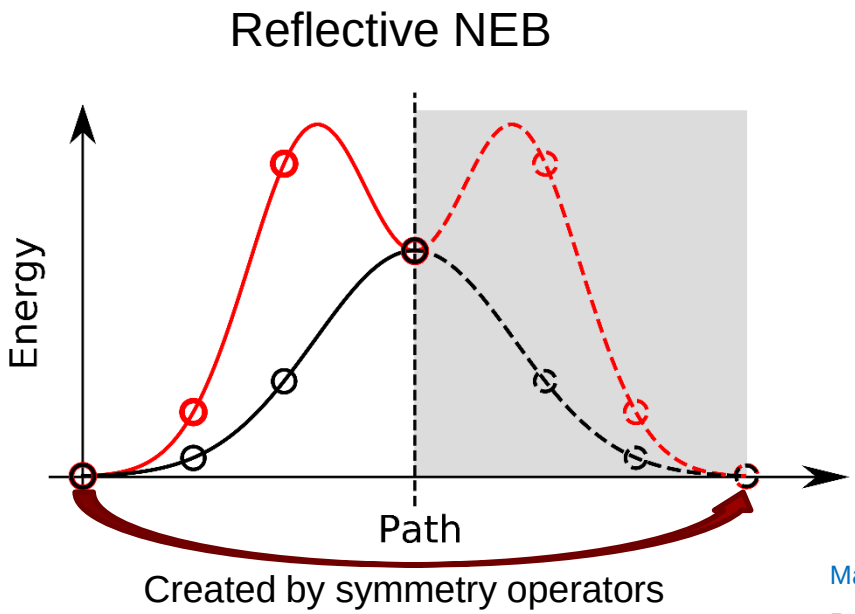

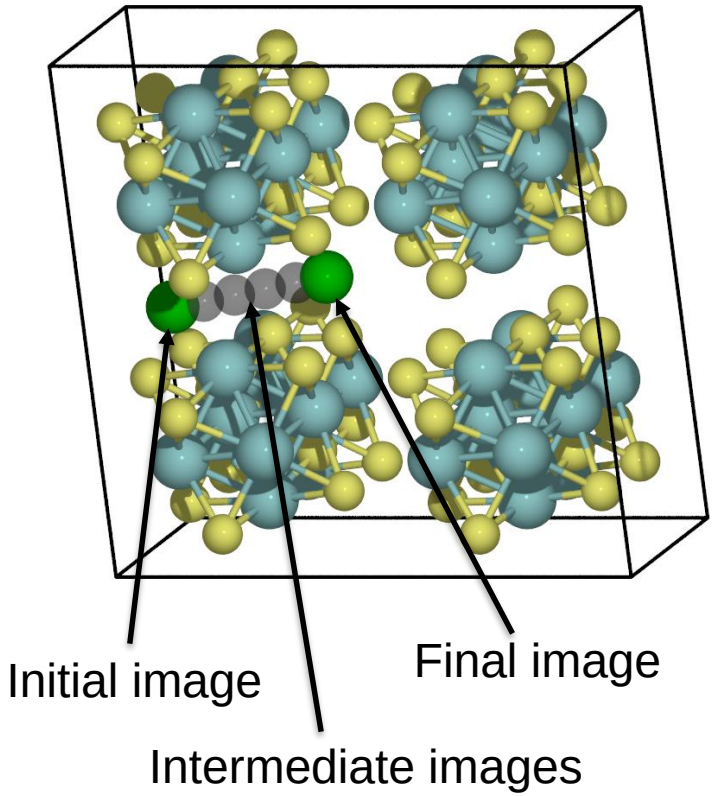

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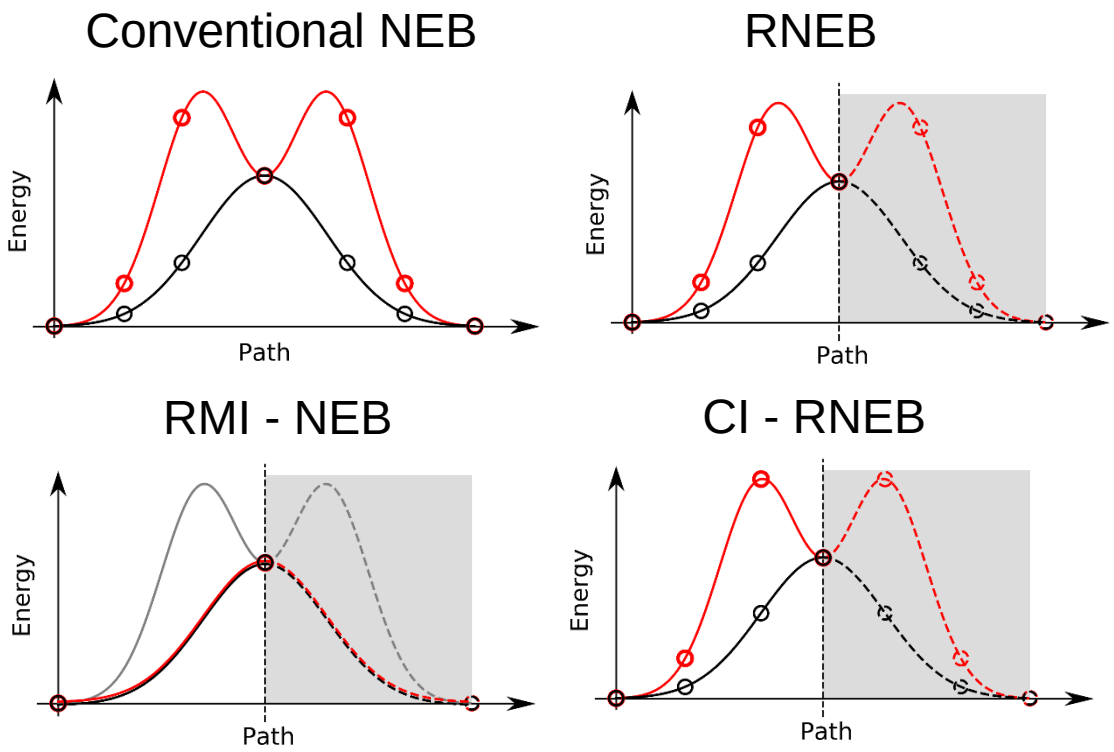
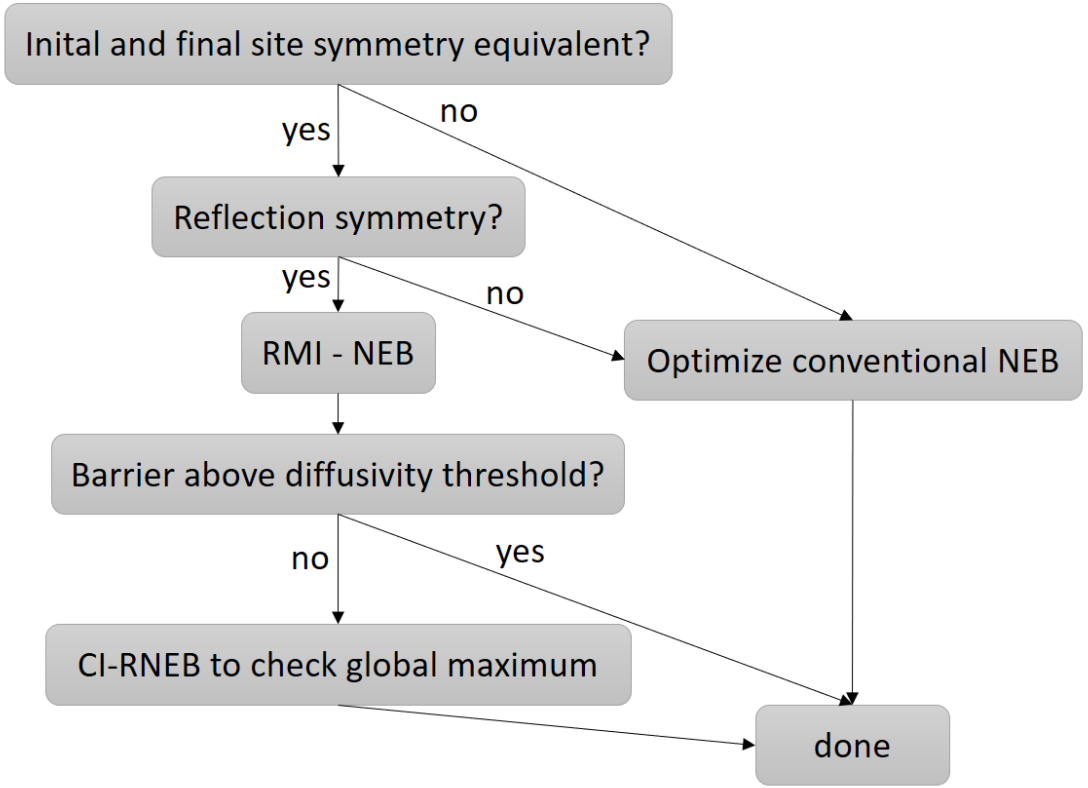
- NEB logic tries to minimize computational cost





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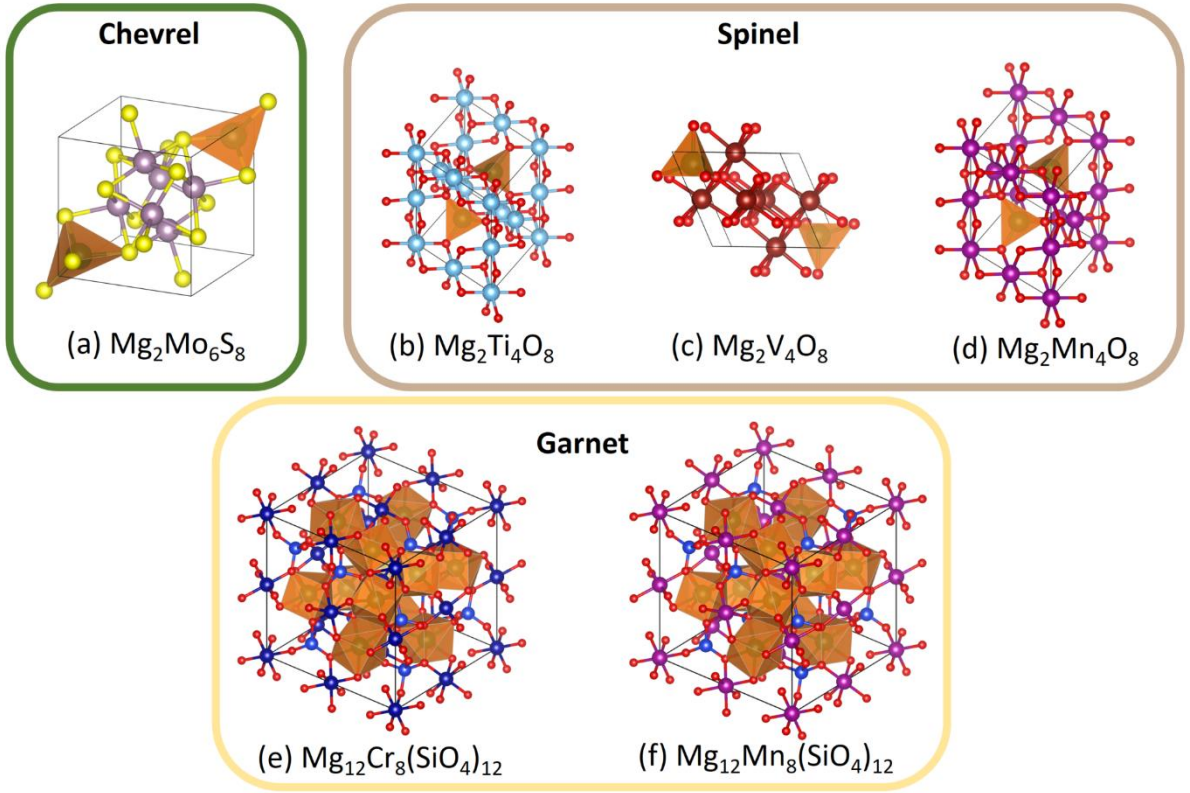
Insertion batteries – Results

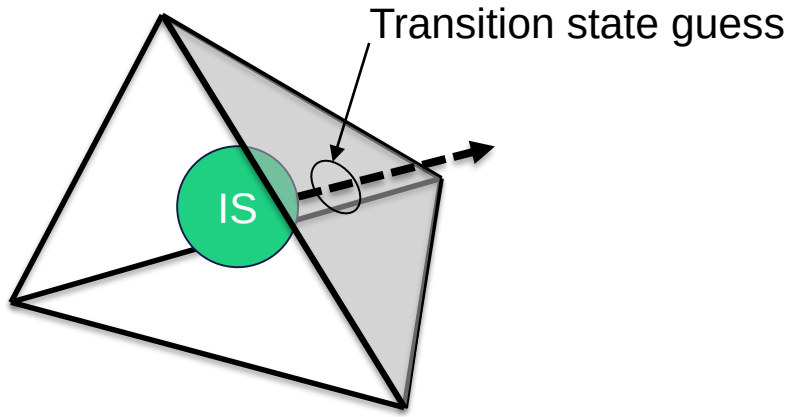
- NEB calculations 3-4 orders of magnitude slower than unit cell relaxation
- Slow (no) diffusion in multivalent batteries
- Systematic study of 16 Mg-battery candidates
  - No partial occupancies
  - Percolating paths exist
  - Metal in cathode does not exceed maximum oxidation state upon fully charging
  - Energy above the hull < 0.5 eV/atom
  - Only reflective-paths are considered (four or less)
- Can we use data to predict the barrier?



Paper III - **Bölle**, Mathiesen, Nielsen, Vegge, García-Lastra, Castelli. *Batteries and Supercaps*.; 3(6), 470-470 (2020).

Insertion batteries – importance of transition state

- NEB calculations 3-4 orders of magnitude slower than unit cell relaxation
- Geometrical features solely based on relaxed unit cell
- Transition state guess shows stronger correlation with kinetic barrier



	Pearson correlation coefficient
Minimum ion-anion$_{NN}$ separation distance initial configuration	**-0.09**
Maximum ion-anion$_{NN}$ separation distance initial configuration	**+ 0.02**
Minimum ion-anion$_{NN}$ separation distance transition state	**- 0.25**
Maximum ion-anion$_{NN}$ separation distance transition state	**-0.38**

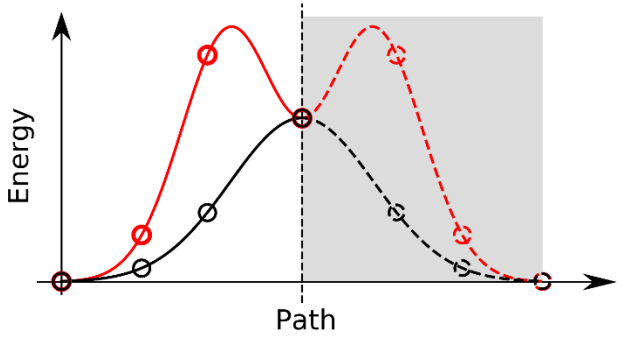
Paper IV - **Bölle**, Vegge, García-Lastra, Castelli. *To be submitted* (2021).

Insertion batteries – NEB logic

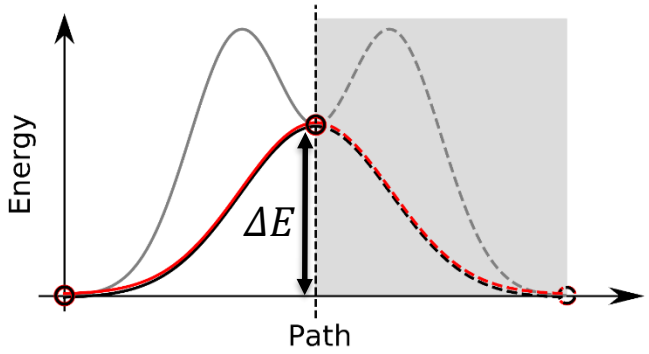
5. Prepare NEB paths

- Reduce the number of images to calculate using reflection symmetry
- A single image is enough to estimate height of barrier
- Climbing image will find global energy maximum (Transition state)

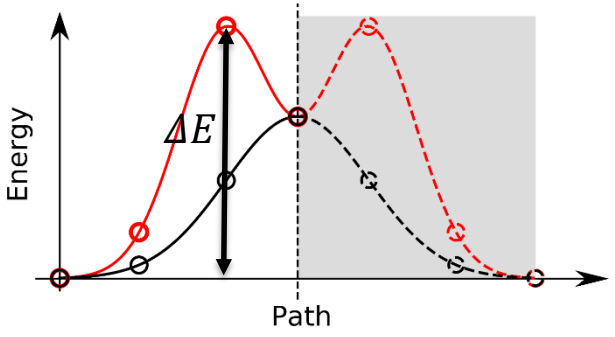
Reflective NEB (RNEB)



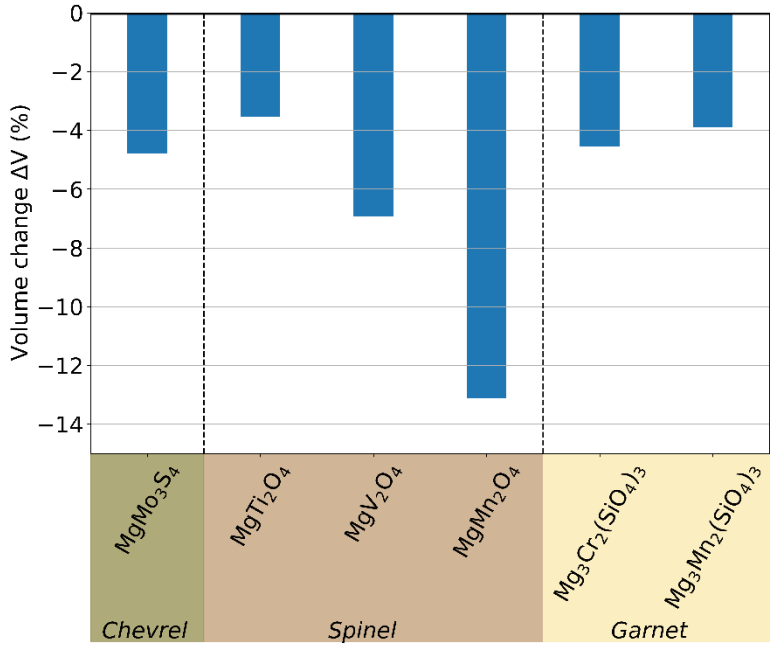
Reflective middle -image NEB

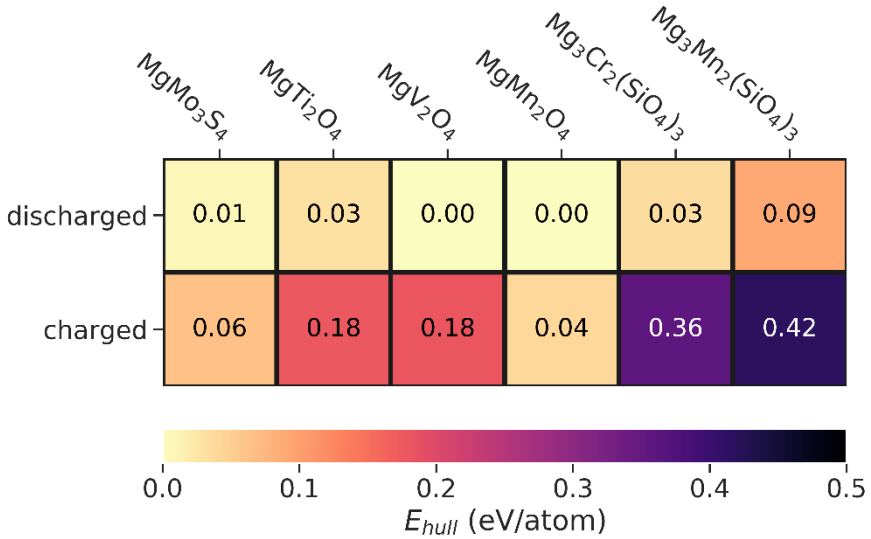


Climbing image - RNEB



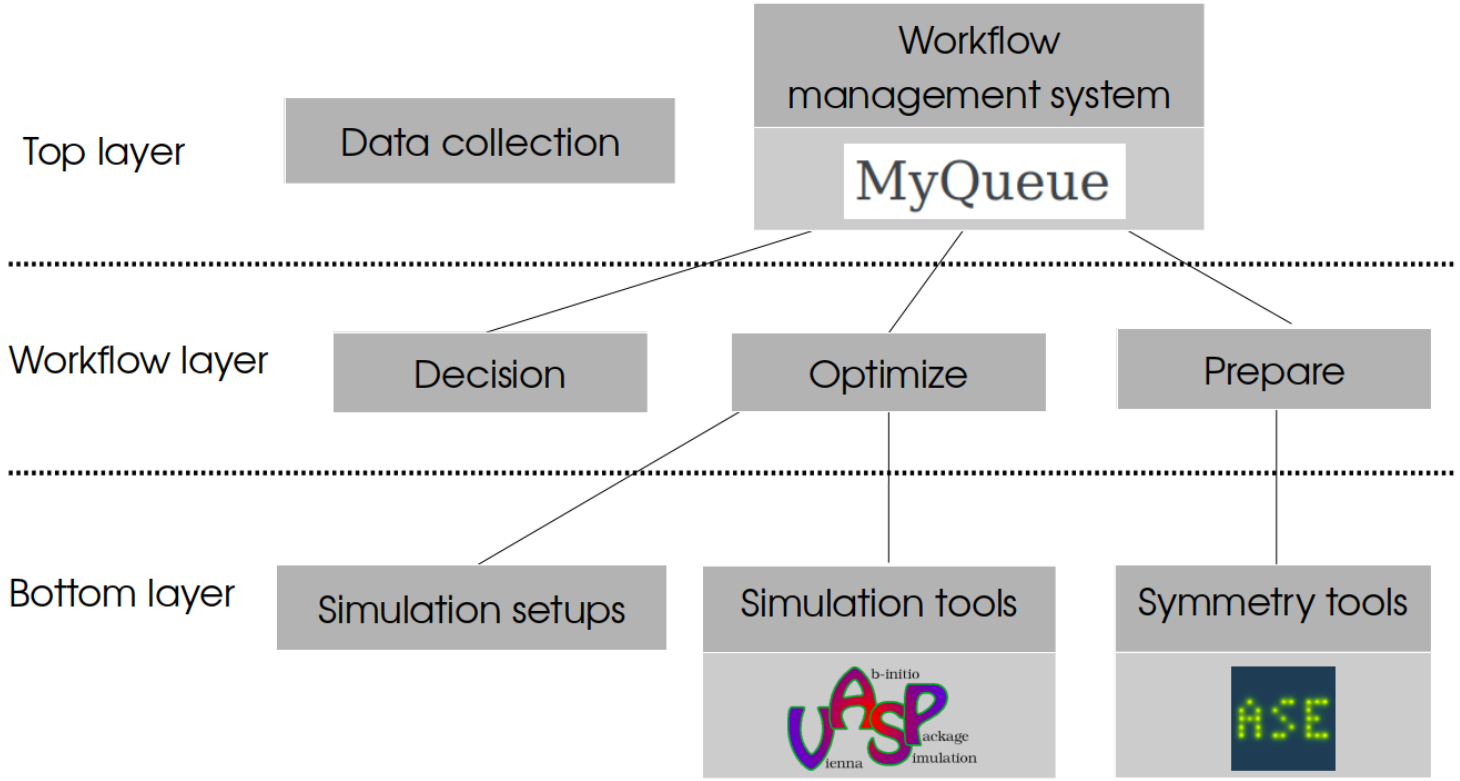
Insertion batteries – volume change and OCVs





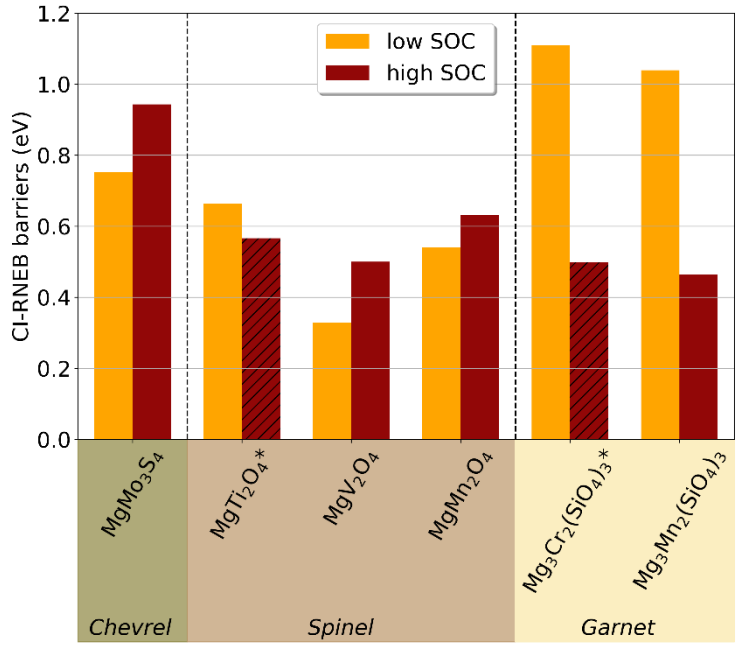
Methods – scientific workflow tools

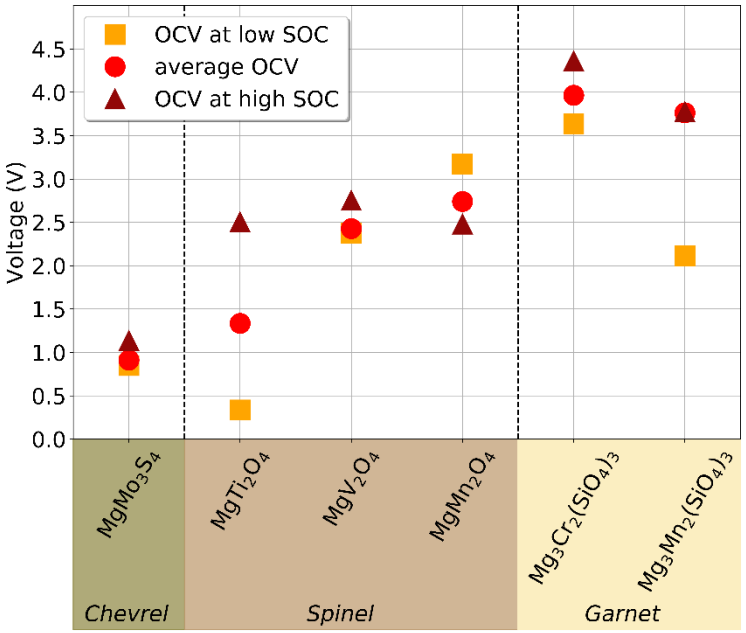
- Use what you already know and make use of existing knowledge
  - "workflow-language"
- Modularize code based on the tools used
  - Exchange VASP code with e.g. GPAW
- Structure tasks to have well defined input and output
  - Reuse and testing



[1] Bölle et al. Batteries & Supercaps 3(6), 488-498 (2020)

Insertion batteries – OCVs and kinetic barriers





Methods – scientific workflow tools

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- DFT-Code specific errors
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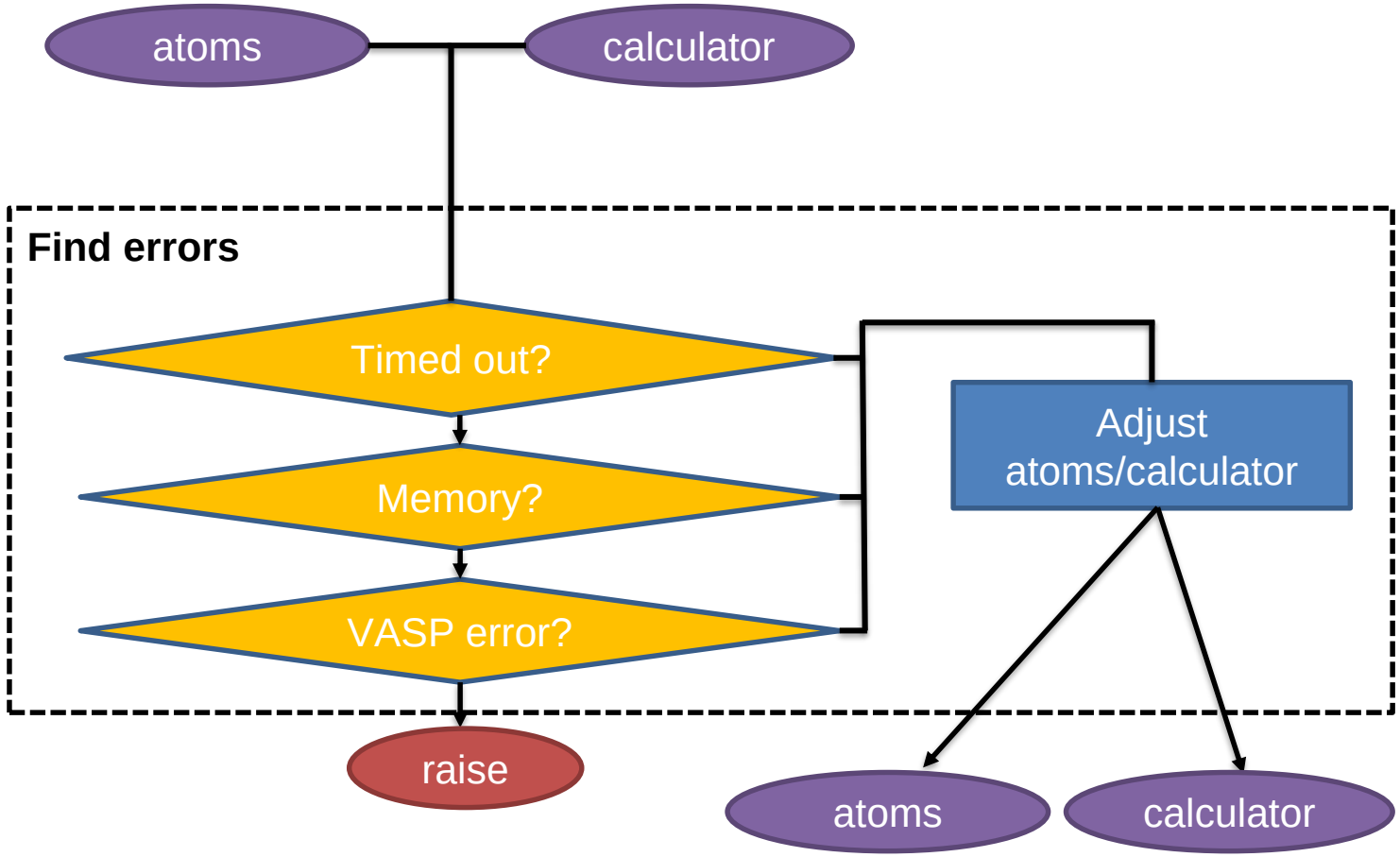

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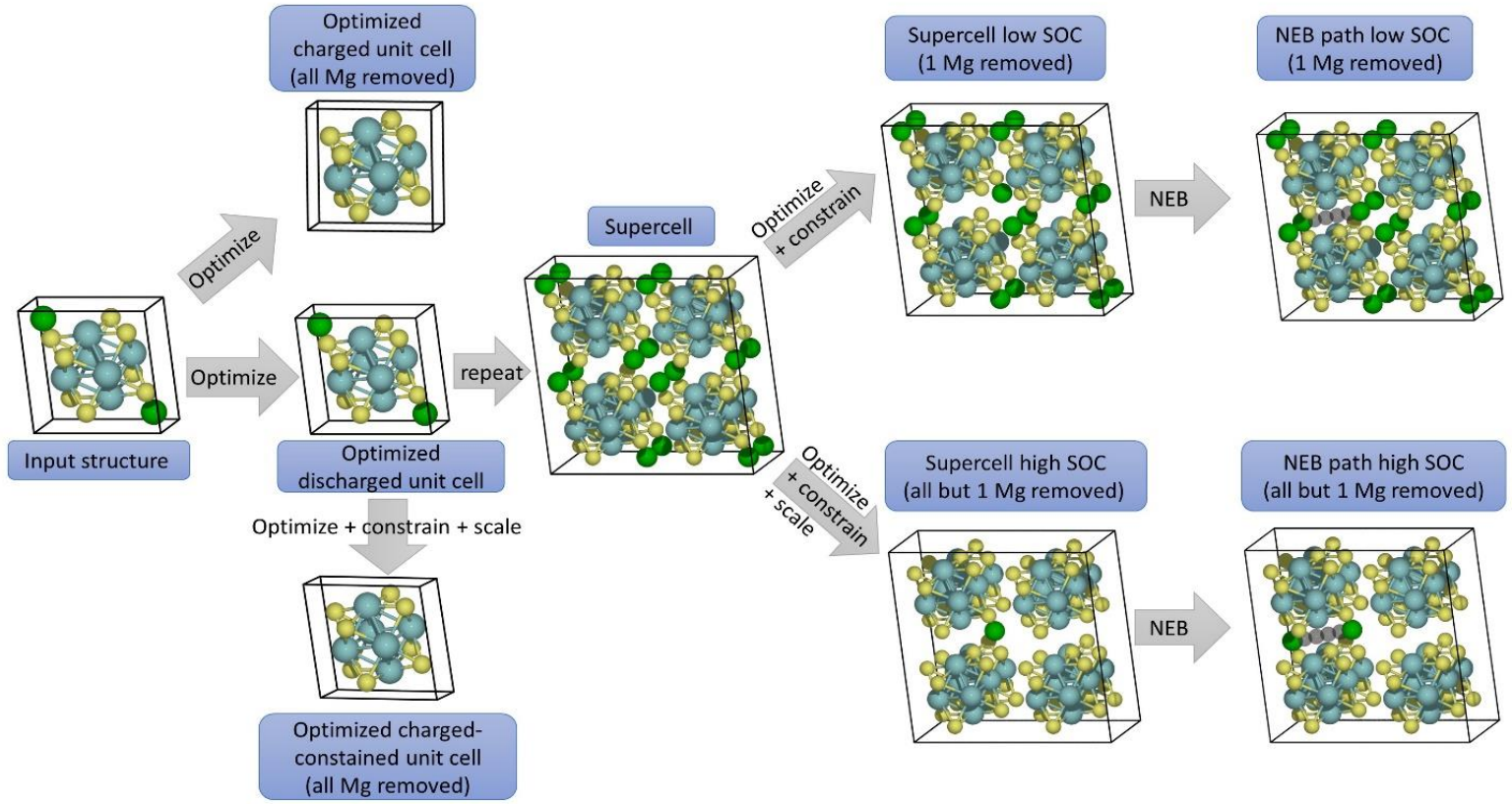
Insertion batteries – workflow complexity

- There are at least 9 unique crystal structures for a single material

- **Optimize** ≙ Optimize cell, volume and atomic positions
- **Optimize + constrain** ≙ Optimize atomic positions
- **Optimize + constrain + scale** ≙ Optimize atomic positions after scaling cell and positions



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