High-throughput computational screening of fast Li-ion conductors

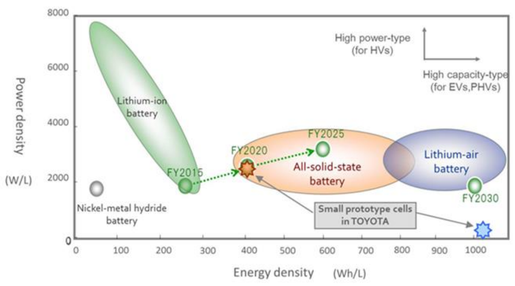
Tushar Thakur, Loris Ercole and Nicola Marzari

THEOS and NCCR-MARVEL, École Polytechnique Fédérale de Lausanne (EPFL), Switzerland

1. Broader Context

I. Motivation

- Conventional electrolytes are volatile and flammable.
- Solid State Electrolytes (SSE) have higher energy density and high operable temperature.

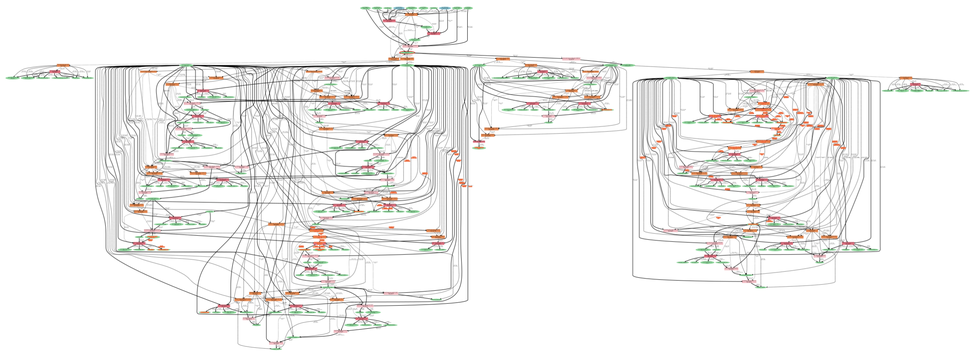


II. Problem

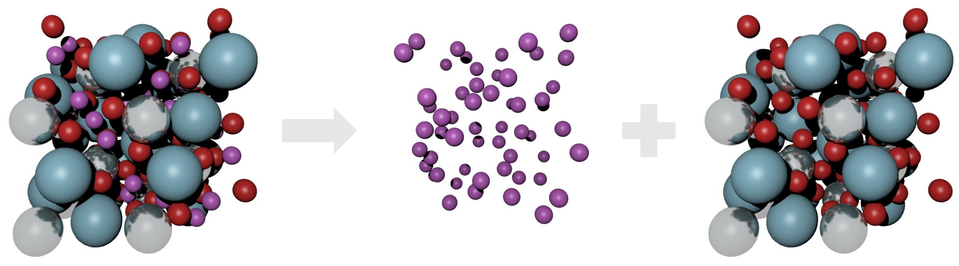
- Experimental approaches for materials discovery is human intensive and not easily scalable, hence the need for computational screening.
- To compute ionic conductivity on hundreds of structures a fast and accurate potential is required.

III. Method

- AiiDA serves as an infrastructure to automate and manage the complex workflows required in this screening [1] [2]. An example provenance -



- Pinball model computes forces to run MD simulations [3].



$$\hat{\mathcal{H}}_1 = \frac{1}{2}\sum_p^P M_p \dot{R}_p^2 + \alpha_{1,2}\int n_{H_0}(\boldsymbol{r}) V_{ext}^P(\boldsymbol{r}) d\boldsymbol{r} + \beta_{1,2} \sum_{I,J}^{H_0-P,P-P} \frac{Z_I Z_J}{|\boldsymbol{R}_I - \boldsymbol{R}_J|}$$

- DFT forces are well reproduced by the pinball model

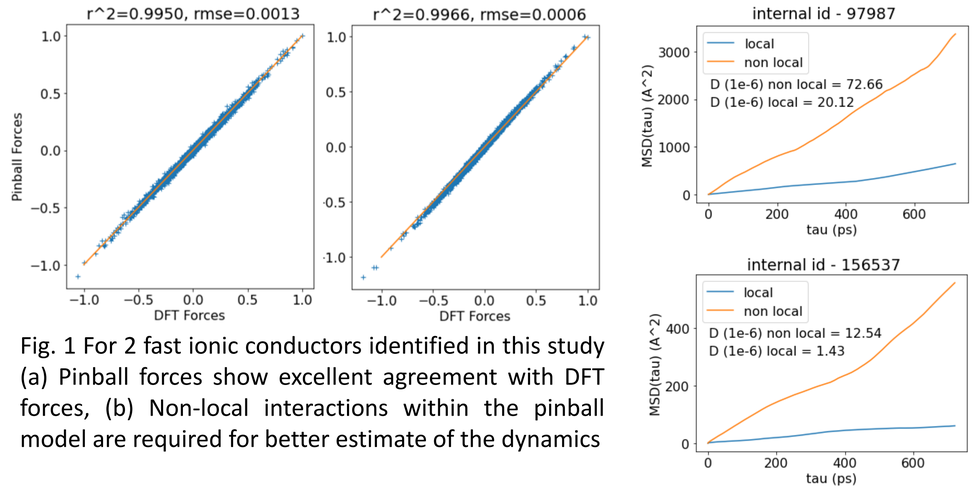
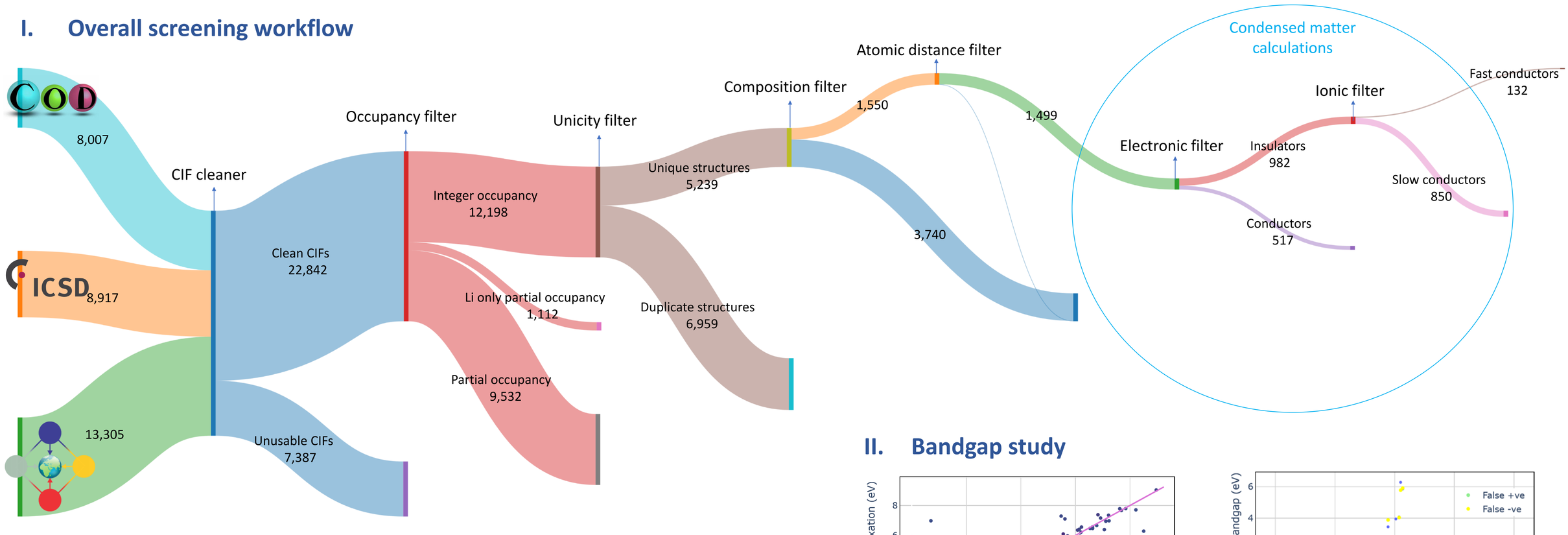


Fig. 1 For 2 fast ionic conductors identified in this study (a) Pinball forces show excellent agreement with DFT forces, (b) Non-local interactions within the pinball model are required for better estimate of the dynamics

2. Results

I. Overall screening workflow



II. Bandgap study

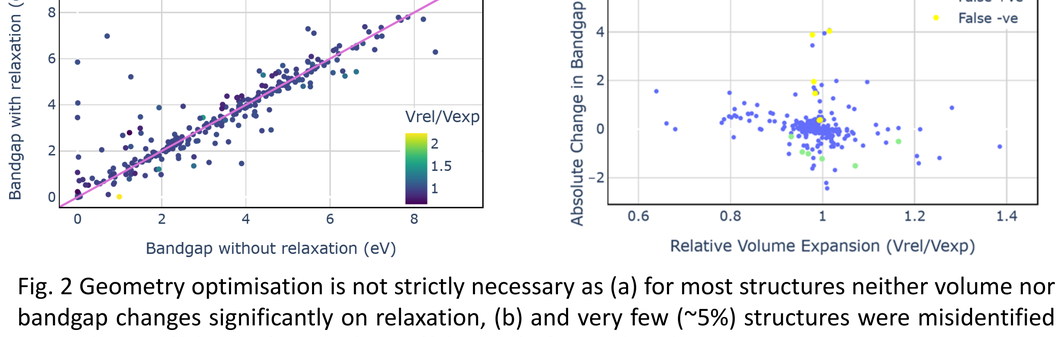


Fig. 2 Geometry optimisation is not strictly necessary as (a) for most structures neither volume nor bandgap changes significantly on relaxation, (b) and very few (~5%) structures were misidentified as conductors (false +ve) or insulators (false –ve), if not relaxed

III. Pinball MD

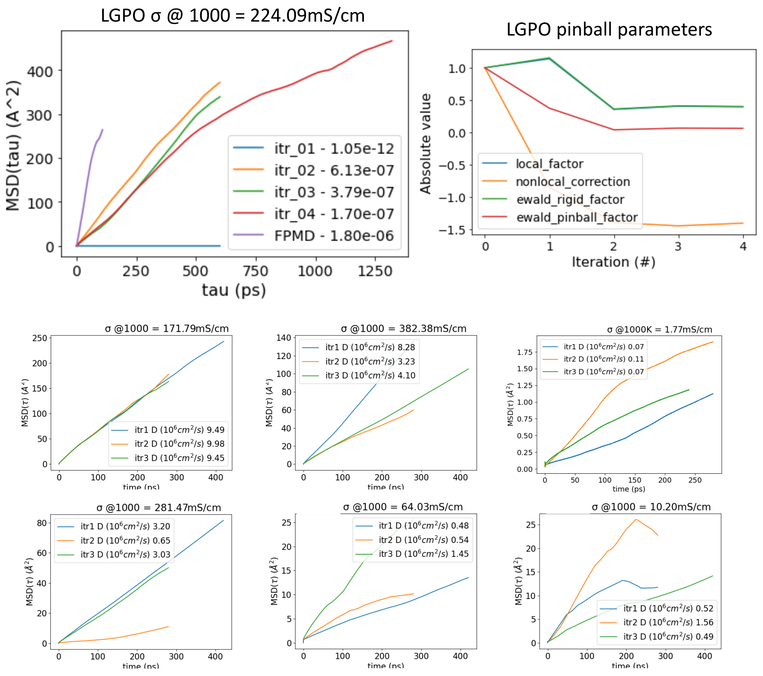


Fig. 3 Self-consistent convergence of MSD plots and pinball parameters at 1000K of a few fast conductors identified in this study

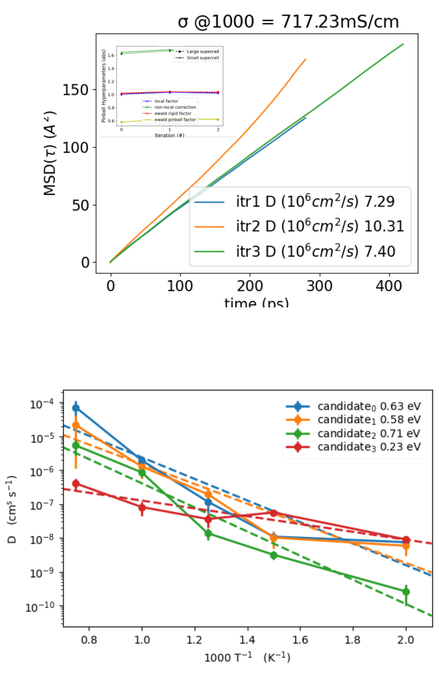


Fig. 4 Diffusion coefficients of a few promising candidates with their activation barriers

3. Conclusions

With this workflow implemented in AiiDA, one can find fast Li-ion conductors making use of the pinball model to run MD calculations that are as precise as *ab initio* MD while being a few hundred times less computationally expensive.

4. References

[1] SP Huber et al – Scientific Data 7, 300 (2020)
[2] T. Thakur *et al* (in preparation)
[3] L Kahle, A Marcolongo, N Marzari - Physical Review Materials 2, 065405 (2018)

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