

Supporting Information: Machine Learning Enabled Computational Screening of Inorganic Solid Electrolytes for Suppression of Dendrite Formation in Lithium Metal Anodes

Zeeshan Ahmad,[†] Tian Xie,[‡] Chinmay Maheshwari,[†] Jeffrey C. Grossman,[‡] and Venkatasubramanian Viswanathan^{*,†,¶}

[†]*Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA*

[‡]*Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

[¶]*Department of Physics, Carnegie Mellon University, Pittsburgh, Pennsylvania 15213, USA*

E-mail: venkvis@cmu.edu

Interface between Li metal and solid electrolyte

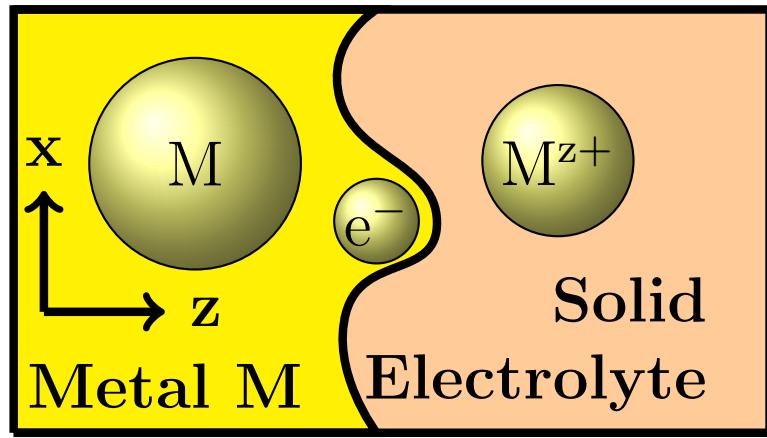


Figure S1: A general 2D interface $z = f(x)$ between Li metal and an inorganic solid electrolyte during electrodeposition.

Details of the machine learning models

Isotropic criteria – Crystal Graph Convolutional Neural Network (CGCNN)

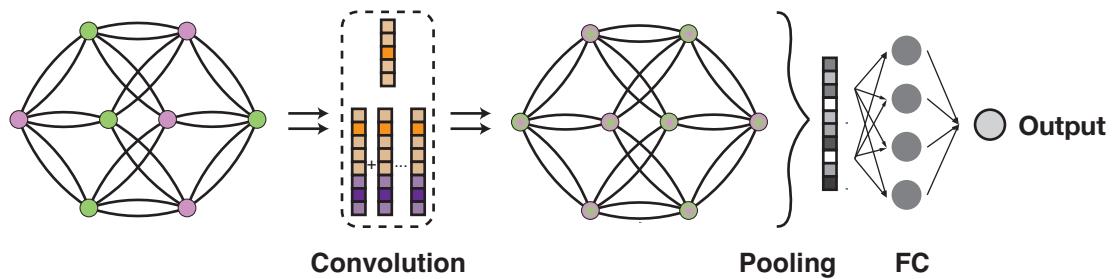


Figure S2: Schematic showing the architecture of CGCNN.



Figure S3: Visualization of the latent space representations of 500 random training and 500 random test crystals using t-distributed stochastic neighbor embedding algorithm for CGCNN.

Table S1: Hyper-parameters of the best performing CGCNN model for predicting shear and bulk modulus.

Property ^a	Shear modulus	Bulk modulus
Learning rate	1.0×10^{-2}	2.0×10^{-2}
Weight decay	1.0×10^{-4}	1.0×10^{-6}
Number of convolutional layers	4	4

^a Each model was trained for 1000 epochs, and the learning rate was reduced by 10 fold after 800 epochs.

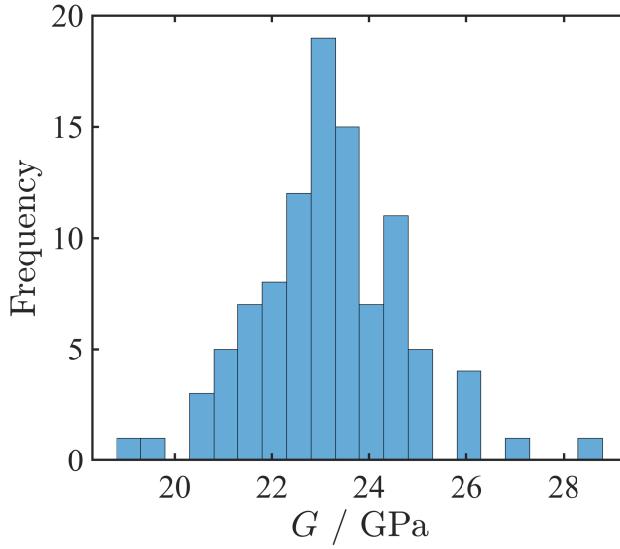


Figure S4: Ensemble of values for shear modulus of Li_5Sn_2 (mp id: mp-30766).

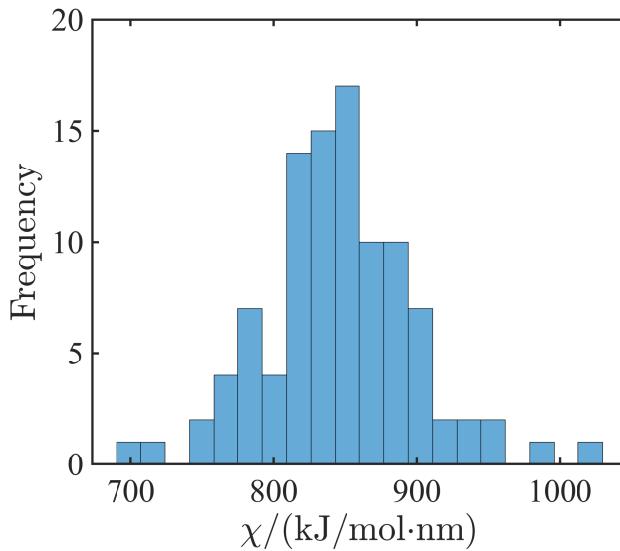


Figure S5: Ensemble of values for the stability parameter of Li_5Sn_2 (mp id: mp-30766).

Anisotropic criteria

List of descriptors used for developing regression model of the elastic tensor. The descriptors have been inspired by Ref. 1 (with some modifications) which used them for ionic conductivity predictions. Some descriptors which were expected to affect only ionic conductivity were removed.

1. Volume per atom (\AA^3)
2. Standard deviation in Li neighbor count
3. Standard deviation in Li bond ionicity
4. Li bond ionicity
5. Li neighbour count
6. Li-Li bonds per Li
7. Sublattice Bond ionicity
8. Sublattice neighbour count
9. Anion framework coordination
10. Minimum anion-anion separation distance (\AA)
11. Volume per anion (\AA^3)
12. Minimum Li-anion separation distance (\AA)
13. Mass density
14. Minimum Li-Li separation distance (\AA)
15. Sublattice Electronegativity
16. Ratio of bond ionicities of Li and sublattice

17. Ratio of neighbor counts of Li and sublattice

The search space for anisotropic solid-solid interfaces involves low index facets of Li and solid electrolyte. The crystallographic directions $\langle 100 \rangle$, $\langle 010 \rangle$, $\langle 001 \rangle$, $\langle 110 \rangle$, $\langle 011 \rangle$, $\langle 101 \rangle$ and $\langle 111 \rangle$ of the solid electrolytes were considered normal to the interface with Li metal anode. We used the elastic tensor of the conventional unit cell obtained from that of the primitive unit cell provided by `materials project`. The new elastic tensor for an arbitrary crystallographic orientation was obtained by rotating the axes and applying the transformation rules.²

Table S2: Available # of Li containing compounds in training data for each crystal class. Tetragonal (I) refers to the point groups 4, $\bar{4}$ and 4/m while (II) refers to point groups 4mm, 422, $\bar{4}2m$ and 4/mmm. Trigonal (I) refers to point groups 3 and $\bar{3}$ while (II) refers to point groups 32, $\bar{3}m$ and 3m.

Crystal class	# of materials	# unique surfaces ^a
Cubic	177	3
Hexagonal	55	5
Orthorhombic	65	7
Monoclinic	63	7
Tetragonal (I)	7	6
Tetragonal (II)	56	6
Trigonal (I)	2	5
Trigonal (II)	44	5
Triclinic	13	7
Total	482	2401

^a out of the low index surfaces.

Using Table S2, one can calculate the total # of interfaces screened over as: # of unique surfaces \times # Li surfaces (DFT) + # of unique surfaces \times # Li surfaces (predicted) = $2401 \times 4 + 548 \times 3 \times 4 = 16180$.

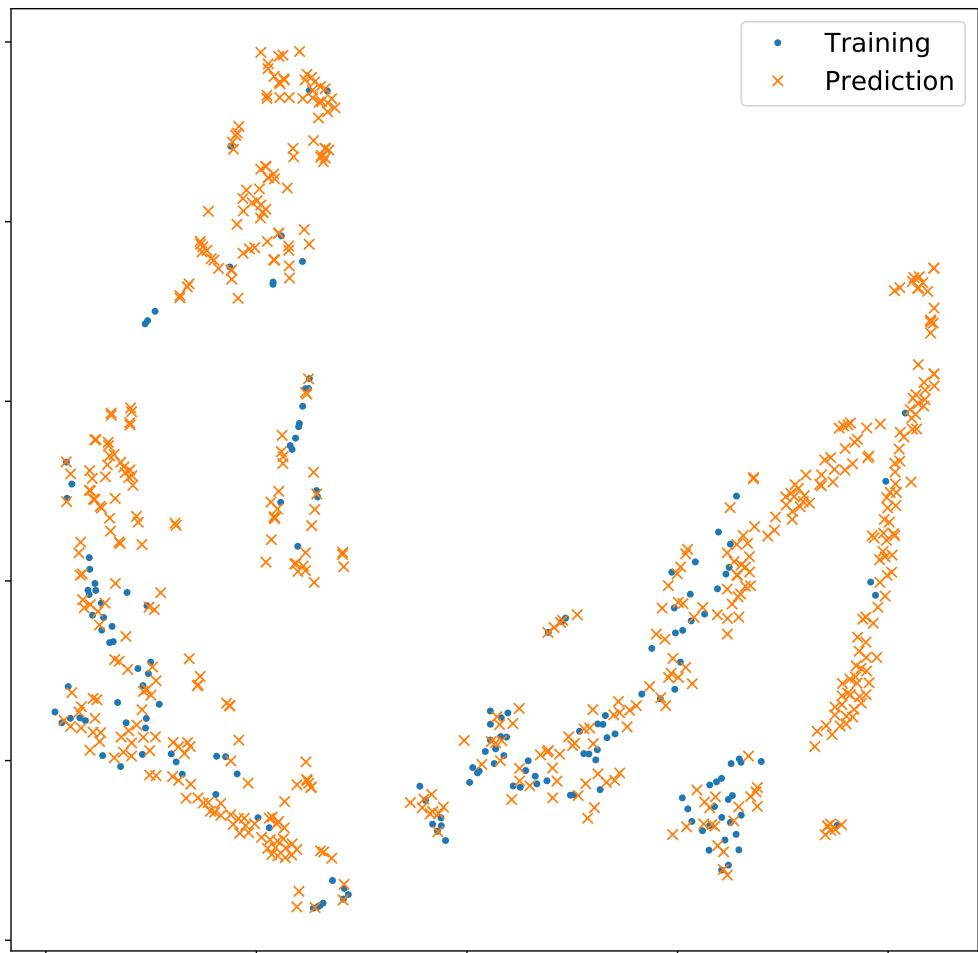


Figure S6: Visualization of the latent space representations of all training and all test crystals using t-distributed stochastic neighbor embedding algorithm for the machine learning models for anisotropic case.

Table S3: Models used for predicting the elastic constants of the cubic crystal class. Hyperparameter optimization was performed using grid-search on **scikit-learn** package.³ The naming of hyperparameters is the same as that in **scikit-learn** package.

Elastic constant	Model	Hyperparameters	
		Parameter	Value
C_{11}	Gradient boost regression	estimators	500
		max depth	20
		learning rate	0.01
		max features	sqrt
		minimum samples per leaf	10
		minimum samples split	2
		subsample	1.0
		α	0.01
C_{12}	Kernel ridge regression	kernel	rbf
		gamma	0.01
		estimators	500
		max depth	3
C_{44}	Gradient boost regression	learning rate	0.01
		max features	auto
		minimum samples per leaf	4
		minimum samples split	4
		subsample	1.0

Table S4: Surface energies used in calculation of stability parameter.⁴

Li surface	Surface Energy (J/m ²)
(1 0 0)	0.46
(1 1 0)	0.50
(1 1 1)	0.54
(2 1 1)	0.54

Calculation of Other Properties of Screened Electrolytes

Calculation of probability of superionic conduction: The probability of superionic conduction is obtained using five atomistic features 6, 7, 9, 12 and 14 from the list above:¹

$$P_{\text{ion}} = \frac{1}{1 + \exp(-\sum_i \theta_i x_i)}$$

$$\sum_i \theta_i x_i = 0.184 \times x_6 - 4.009 \times x_7 - 0.467 \times x_9 + 8.699 \times x_{12} - 2.170 \times x_{14} - 6.564$$

Here x_i are the values of atomistic feature i without any scaling/normalization.

Calculation of corrected band gap: One can correct for DFT band gap through an empirical scaling relation proposed by Morales-García et al.⁵:

$$E_{\text{gap}}(G_0W_0) = 1.358E_{\text{gap}}(\text{PBE}) + 0.904$$

$$E_{\text{gap}}(\text{exp}) = 0.998E_{\text{gap}}(G_0W_0) + 0.014$$

Data Files

- shear modulus of 60,648 compounds predicted using CGCNN: shear-modulus.xlsx
- bulk modulus of 60,648 compounds predicted using CGCNN: bulk-modulus.xlsx
- Anisotropic stability parameter for training data: train-chi.xlsx
- Anisotropic stability parameter for predicted data: pred-chi.xlsx

References

- (1) Sendek, A. D.; Yang, Q.; Cubuk, E. D.; Duerloo, K.-A. N.; Cui, Y.; Reed, E. J. Holistic computational structure screening of more than 12000 candidates for solid lithium-ion conductor materials. *Energy Environ. Sci.* **2017**, *10*, 306–320.
- (2) Ahmad, Z.; Viswanathan, V. Role of anisotropy in determining stability of electrodeposition at solid-solid interfaces. *Phys. Rev. Materials* **2017**, *1*, 055403, DOI: 10.1103/PhysRevMaterials.1.055403.
- (3) Pedregosa, F. et al. Scikit-learn: Machine Learning in Python. *J. Mach. Learn. Res.* **2011**, *12*, 2825–2830.
- (4) Jain, A.; Ong, S. P.; Hautier, G.; Chen, W.; Richards, W. D.; Dacek, S.; Cholia, S.; Gunter, D.; Skinner, D.; Ceder, G.; Persson, K. A. The Materials Project: A materials genome approach to accelerating materials innovation. *APL Mater.* **2013**, *1*, 011002, DOI: 10.1063/1.4812323.
- (5) Morales-García, A.; Valero, R.; Illas, F. An Empirical, yet Practical Way To Predict the Band Gap in Solids by Using Density Functional Band Structure Calculations. *The Journal of Physical Chemistry C* **2017**, *121*, 18862–18866, DOI: 10.1021/acs.jpcc.7b07421.