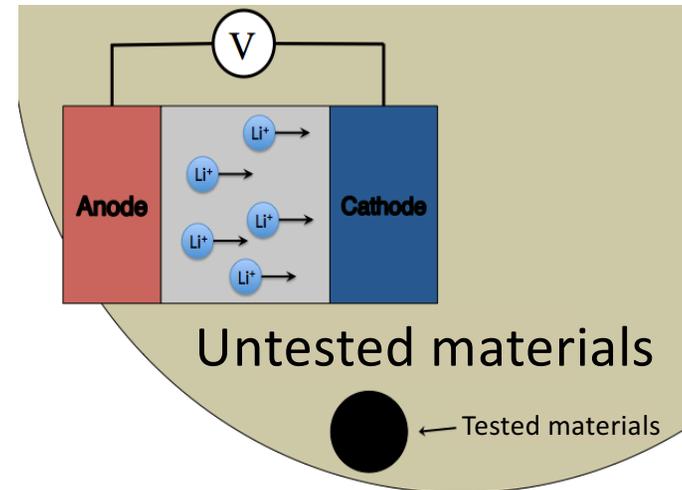
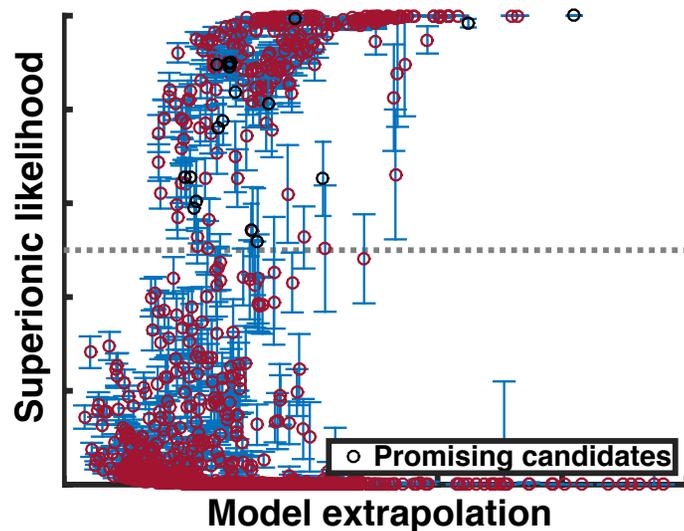


# Designing a better battery with machine learning



Austin D. Sendek, Ekin D. Cubuk, Qian Yang, Gowoon Cheon, Evan R. Antoniuik, Karel-Alexander N. Duerloo, Yi Cui, Evan J. Reed  
MATLAB Expo 2017

# Lithium-ion batteries will enable major innovations in energy



Automotive



Renewable grid



Drones/aviation



Mobile electronics

# Today's lithium-ion batteries still face many challenges

Safety

Energy density

Cost

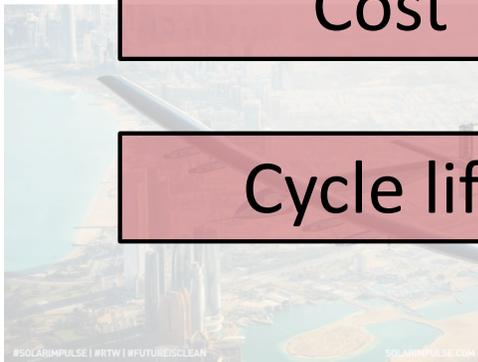
Cycle life



Renewable grid

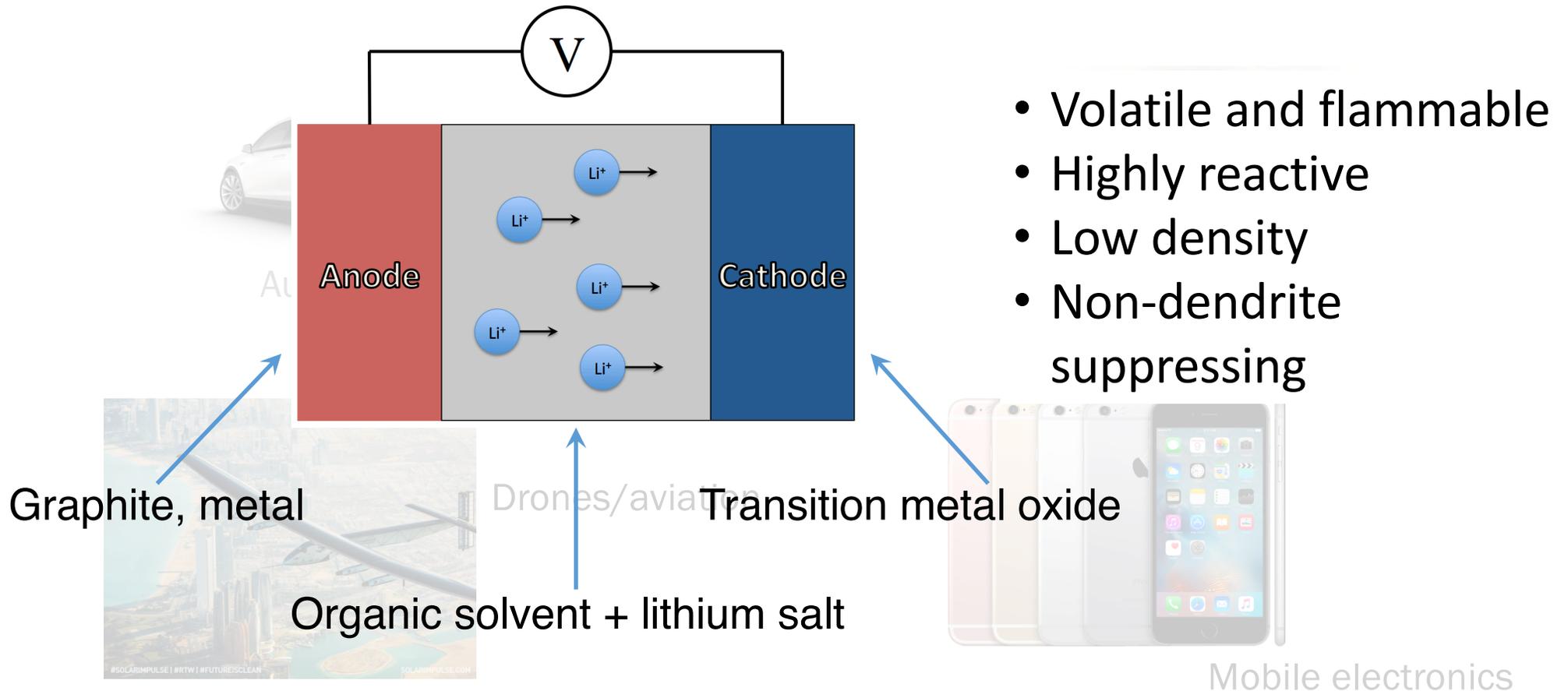


Mobile electronics

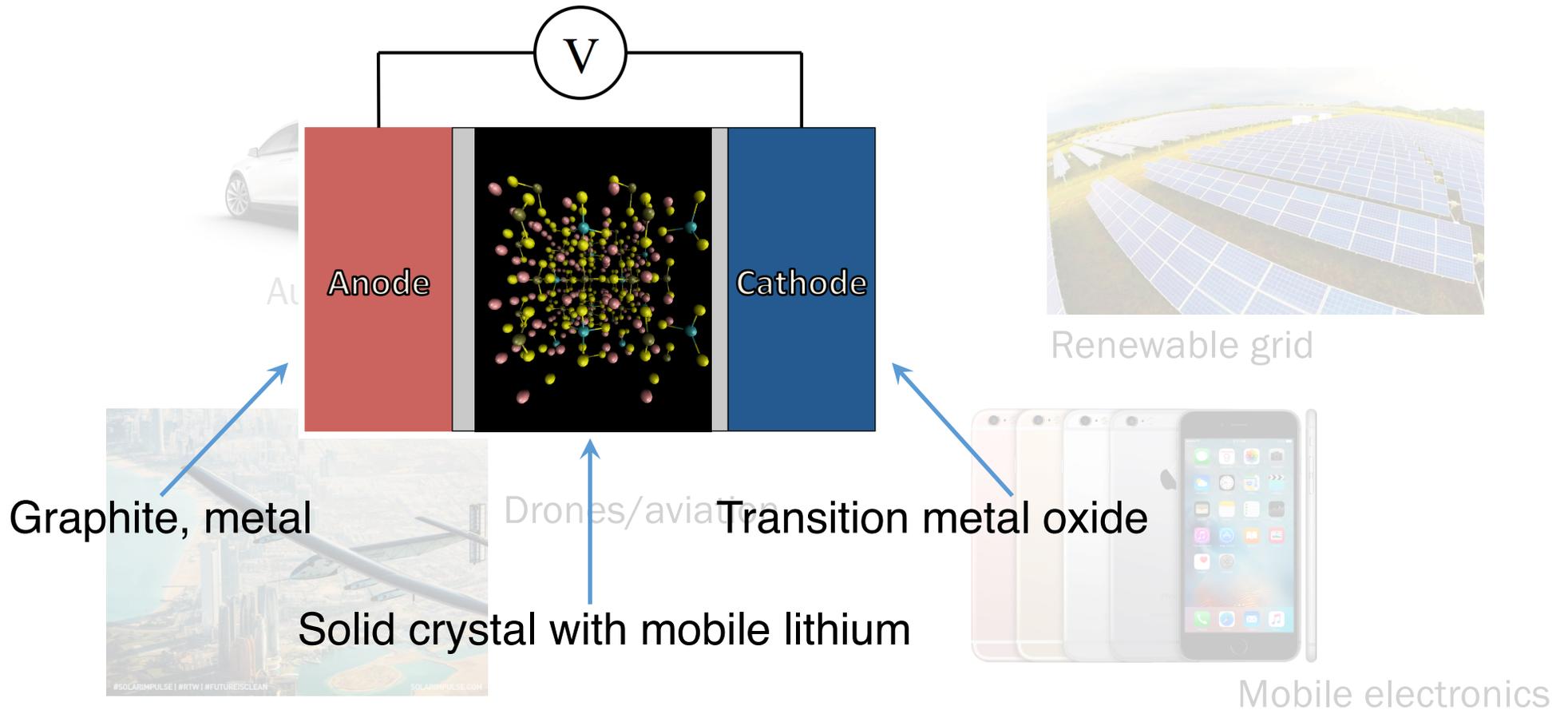


Drones/aviation

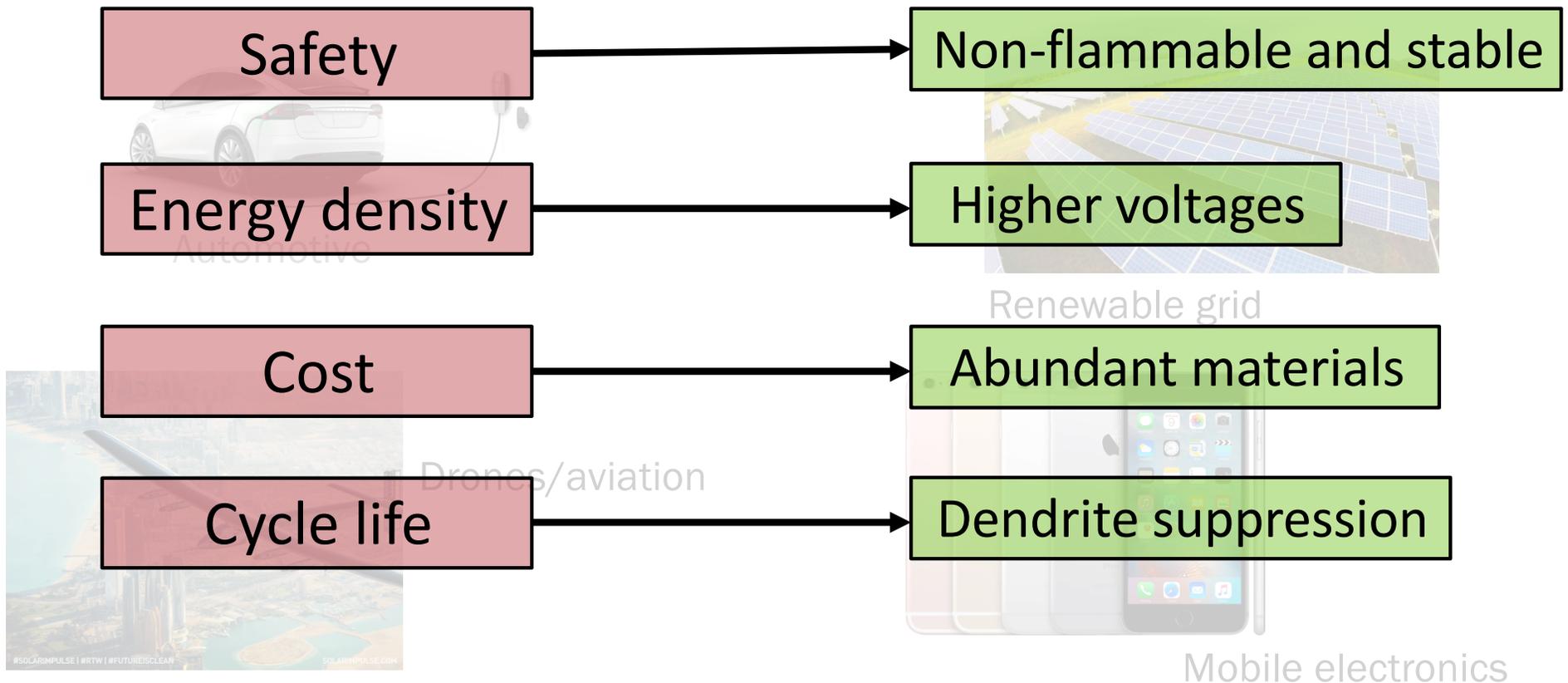
## A major problem worth solving: liquid electrolytes



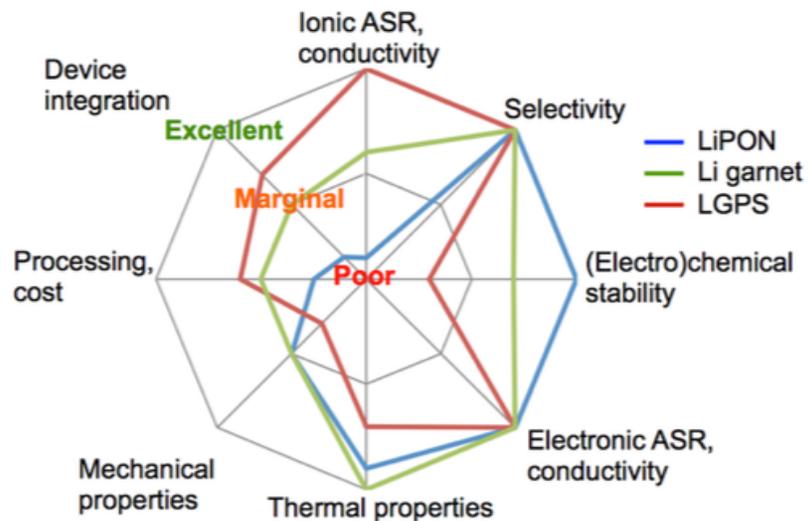
# A major problem worth solving: liquid electrolytes



# Solid electrolytes represent a promising route to improvement



# The challenge: trial-and-error search for candidates satisfying many criteria



Source: US Department of Energy Advanced Research Projects Agency – Energy

*Electrochimica Acta*, 1977, Vol. 22, pp. 773–781. Pergamon Press. Printed in Great Britain

### RECENT RESULTS ON LITHIUM ION CONDUCTORS

ROBERT A. HUGGINS  
Center for Materials Research, Stanford University,  
Stanford, California 94305, U.S.A.

(Received 1 September 1976)

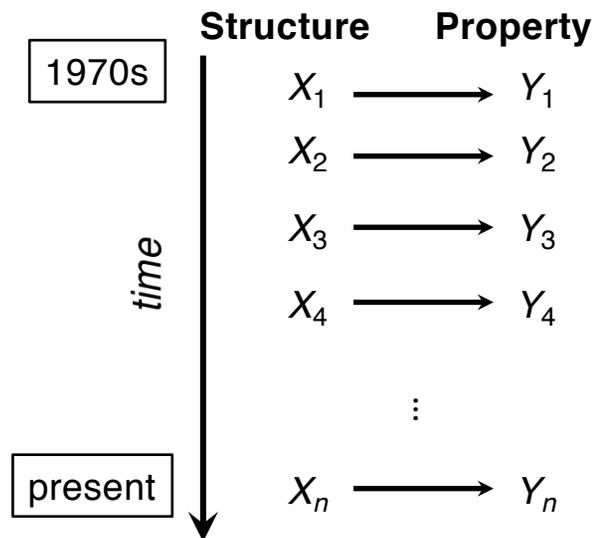
**LETTERS**  
PUBLISHED ONLINE: 31 JULY 2011 | DOI: 10.1038/NMAT3066

nature  
materials

### A lithium superionic conductor

Noriaki Kamaya<sup>1</sup>, Kenji Homma<sup>1</sup>, Yuichiro Yamakawa<sup>1</sup>, Masaaki Hirayama<sup>1</sup>, Ryoji Kanno<sup>1\*</sup>, Masao Yonemura<sup>2</sup>, Takashi Kamiyama<sup>2</sup>, Yuki Kato<sup>3</sup>, Shigenori Hama<sup>3</sup>, Koji Kawamoto<sup>3</sup> and Akio Mitsui<sup>4</sup>

## Existing discovery efforts are driven by trial-and-error



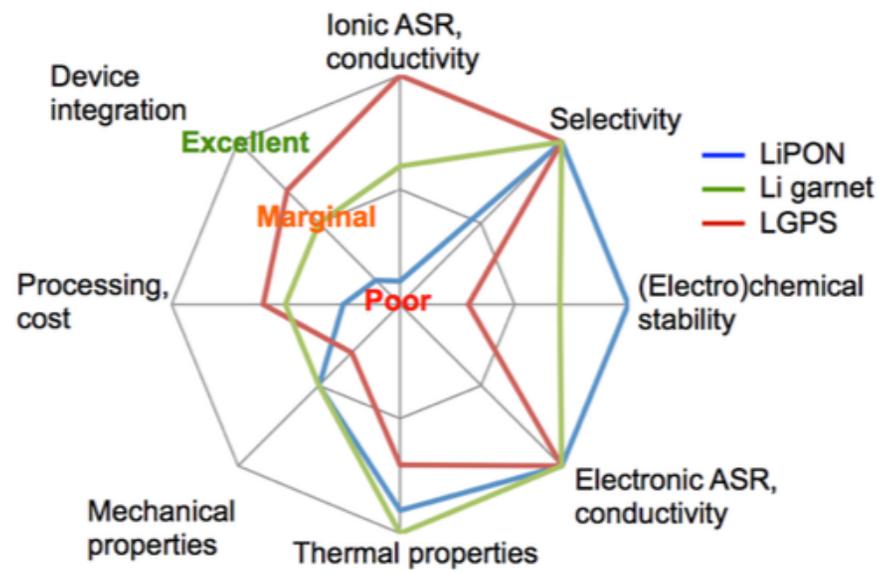
This can be formulated as a **supervised learning** problem:

$$f(X) = Y$$

$$f(\text{structure}) = \text{property}$$

*Can we learn effectively from the data we've already generated?*

Machine learning may improve success rate beyond trial-and-error!



## We draw on the wisdom in the literature for “hypotheses”

Composition	RT bulk ionic conductivity (S cm <sup>-1</sup> )
LiLa(TiO <sub>3</sub> ) <sub>2</sub>	1 × 10 <sup>-3</sup>
Li <sub>9.81</sub> Sn <sub>0.81</sub> P <sub>2.19</sub> S <sub>12</sub>	5.5 × 10 <sup>-3</sup>
Li <sub>10</sub> Ge(PS <sub>6</sub> ) <sub>2</sub>	1.4 × 10 <sup>-2</sup>
Li <sub>10.35</sub> Si <sub>1.35</sub> P <sub>1.65</sub> S <sub>12</sub>	6.5 × 10 <sup>-3</sup>
Li <sub>14</sub> ZnGe <sub>4</sub> O <sub>16</sub> (2)	1.0 × 10 <sup>-6</sup>
Li <sub>2</sub> Ca(NH) <sub>2</sub>	6.4 × 10 <sup>-6</sup>
Li <sub>2</sub> Ge <sub>7</sub> O <sub>15</sub>	5.0 × 10 <sup>-6</sup>
Li <sub>2</sub> NH	2.5 × 10 <sup>-4</sup>
Li <sub>2</sub> S	1.0 × 10 <sup>-13</sup>
Li <sub>13.6</sub> Si <sub>2.8</sub> S <sub>1.2</sub> O <sub>16</sub>	6.0 × 10 <sup>-7</sup>
Li <sub>14</sub> Ge <sub>3</sub> V <sub>2</sub> O <sub>16</sub>	7.0 × 10 <sup>-5</sup>
Li <sub>15</sub> Ge <sub>3</sub> V <sub>2</sub> O <sub>4</sub>	6.03 × 10 <sup>-6</sup>
Li <sub>14.8</sub> Ge <sub>3.4</sub> W <sub>0.6</sub> O <sub>4</sub>	4.0 × 10 <sup>-5</sup>
Li <sub>3</sub> Fe <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	1.0 × 10 <sup>-7</sup>
Li <sub>3</sub> N	5.75 × 10 <sup>-4</sup>
Li <sub>3</sub> P	1.0 × 10 <sup>-3</sup>
γ-Li <sub>3</sub> PS <sub>4</sub>	3.0 × 10 <sup>-7</sup>
Li <sub>3</sub> Sc <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	1.0 × 10 <sup>-10</sup>
β <sub>1</sub> -Li <sub>3</sub> VO <sub>4</sub>	4.4 × 10 <sup>-8</sup>
Li <sub>4</sub> B <sub>7</sub> O <sub>12</sub> Cl	1.0 × 10 <sup>-7</sup>
Li <sub>4</sub> BN <sub>3</sub> H <sub>10</sub>	2.0 × 10 <sup>-4</sup>
γ-Li <sub>4</sub> GeO <sub>4</sub>	3.1 × 10 <sup>-12</sup>
Li <sub>4</sub> SiO <sub>4</sub>	2.4 × 10 <sup>-10</sup>
Li <sub>5</sub> La <sub>3</sub> Bi <sub>2</sub> O <sub>12</sub>	2.0 × 10 <sup>-5</sup>
Li <sub>5</sub> La <sub>3</sub> Nb <sub>2</sub> O <sub>12</sub>	8.0 × 10 <sup>-6</sup>
Li <sub>5</sub> La <sub>3</sub> Ta <sub>2</sub> O <sub>12</sub>	1.5 × 10 <sup>-6</sup>
Li <sub>5</sub> Ni <sub>2</sub>	1.5 × 10 <sup>-7</sup>
Li <sub>6</sub> BaLa <sub>2</sub> Ta <sub>2</sub> O <sub>12</sub>	4.0 × 10 <sup>-5</sup>
Li <sub>6</sub> FeCl <sub>8</sub>	1.0 × 10 <sup>-4</sup>
Li <sub>6</sub> NBr <sub>3</sub>	1.5 × 10 <sup>-7</sup>
Li <sub>6</sub> SrLa <sub>2</sub> Ta <sub>2</sub> O <sub>12</sub>	7.0 × 10 <sup>-6</sup>
Li <sub>7</sub> La <sub>3</sub> Zr <sub>2</sub> O <sub>12</sub>	3.5 × 10 <sup>-4</sup>
Li <sub>7</sub> P <sub>3</sub> S <sub>11</sub>	4.1 × 10 <sup>-3</sup>
LiAlH <sub>4</sub>	2.0 × 10 <sup>-9</sup>
LiAlSiO <sub>4</sub>	1.4 × 10 <sup>-5</sup>
LiBH <sub>4</sub>	2.0 × 10 <sup>-8</sup>
LiI	1.0 × 10 <sup>-6</sup>
LiNH <sub>2</sub>	4.0 × 10 <sup>-10</sup>
α'-LiZr <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	5.0 × 10 <sup>-8</sup>

Feature	Pearson correlation coefficient	
1	Volume per atom <sup>a</sup>	0.20
2	Standard deviation in Li neighbour count	0.22
3	Standard deviation in Li bond ionicity	-0.04
4	Li bond ionicity <sup>a</sup>	-0.18
5	Li neighbour count <sup>a</sup>	-0.19
6	Li-Li bonds per Li <sup>a</sup>	0.06
7	Bond ionicity of sublattice <sup>a</sup>	-0.28
8	Sublattice neighbour count <sup>a</sup>	-0.13
9	Anion framework coordination <sup>a</sup>	-0.06
10	Minimum anion-anion separation distance <sup>a</sup> (Å)	0.09
11	Volume per anion (Å <sup>3</sup> )	-0.01
12	Minimum Li-anion separation distance <sup>a</sup> (Å)	0.20
13	Minimum Li-Li separation distance <sup>a</sup> (Å)	-0.10
14	Electronegativity of sublattice <sup>a</sup>	-0.16
15	Packing fraction of full crystal	0.16
16	Packing fraction of sublattice	0.19
17	Straight-line path width <sup>a</sup> (Å)	0.07
18	Straight-line path electronegativity <sup>a</sup>	-0.29
19	Ratio of features (4) and (7)	-0.03
20	Ratio of features (5) and (8)	-0.18
	Constant term	—

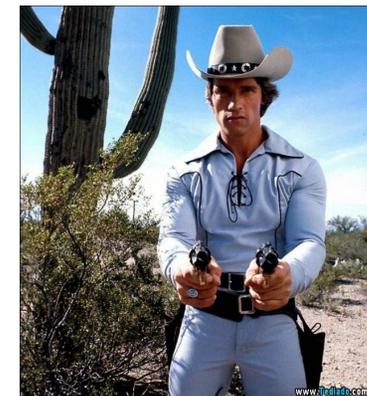
No single feature has strong correlation with ionic conductivity across the broad spectrum of 40 materials

$$C = \text{corr}(X, Y)$$

## We collect 40 experimental measurements of conductivity for solids

Composition	RT bulk ionic conductivity (S cm <sup>-1</sup> )
LiLa(TiO <sub>3</sub> ) <sub>2</sub>	1 × 10 <sup>-3</sup>
Li <sub>9.81</sub> Sn <sub>0.81</sub> P <sub>2.19</sub> S <sub>12</sub>	5.5 × 10 <sup>-3</sup>
Li <sub>10</sub> Ge(PS <sub>6</sub> ) <sub>2</sub>	1.4 × 10 <sup>-2</sup>
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Li <sub>14</sub> ZnGe <sub>4</sub> O <sub>16</sub> (2)	1.0 × 10 <sup>-6</sup>
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Li <sub>14</sub> Ge <sub>2</sub> V <sub>2</sub> O <sub>16</sub>	7.0 × 10 <sup>-5</sup>
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Li <sub>14.8</sub> Ge <sub>3.4</sub> W <sub>0.6</sub> O <sub>4</sub>	4.0 × 10 <sup>-5</sup>
Li <sub>3</sub> Fe <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	1.0 × 10 <sup>-7</sup>
Li <sub>3</sub> N	5.75 × 10 <sup>-4</sup>
Li <sub>3</sub> P	1.0 × 10 <sup>-3</sup>
γ-Li <sub>3</sub> PS <sub>4</sub>	3.0 × 10 <sup>-7</sup>
Li <sub>3</sub> Sc <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	1.0 × 10 <sup>-10</sup>
β <sub>11</sub> -Li <sub>3</sub> VO <sub>4</sub>	4.4 × 10 <sup>-8</sup>
Li <sub>4</sub> B <sub>7</sub> O <sub>12</sub> Cl	1.0 × 10 <sup>-7</sup>
Li <sub>4</sub> BN <sub>3</sub> H <sub>10</sub>	2.0 × 10 <sup>-4</sup>
γ-Li <sub>4</sub> GeO <sub>4</sub>	3.1 × 10 <sup>-12</sup>
Li <sub>4</sub> SiO <sub>4</sub>	2.4 × 10 <sup>-10</sup>
Li <sub>5</sub> La <sub>3</sub> Bi <sub>2</sub> O <sub>12</sub>	2.0 × 10 <sup>-5</sup>
Li <sub>5</sub> La <sub>3</sub> Nb <sub>2</sub> O <sub>12</sub>	8.0 × 10 <sup>-6</sup>
Li <sub>5</sub> La <sub>3</sub> Ta <sub>2</sub> O <sub>12</sub>	1.5 × 10 <sup>-6</sup>
Li <sub>5</sub> Ni <sub>2</sub>	1.5 × 10 <sup>-7</sup>
Li <sub>6</sub> BaLa <sub>2</sub> Ta <sub>2</sub> O <sub>12</sub>	4.0 × 10 <sup>-5</sup>
Li <sub>6</sub> FeCl <sub>8</sub>	1.0 × 10 <sup>-4</sup>
Li <sub>6</sub> NBr <sub>3</sub>	1.5 × 10 <sup>-7</sup>
Li <sub>6</sub> SrLa <sub>2</sub> Ta <sub>2</sub> O <sub>12</sub>	7.0 × 10 <sup>-6</sup>
Li <sub>7</sub> La <sub>3</sub> Zr <sub>2</sub> O <sub>12</sub>	3.5 × 10 <sup>-4</sup>
Li <sub>7</sub> P <sub>3</sub> S <sub>11</sub>	4.1 × 10 <sup>-3</sup>
LiAlH <sub>4</sub>	2.0 × 10 <sup>-9</sup>
LiAlSiO <sub>4</sub>	1.4 × 10 <sup>-5</sup>
LiBH <sub>4</sub>	2.0 × 10 <sup>-8</sup>
LiI	1.0 × 10 <sup>-6</sup>
LiNH <sub>2</sub>	4.0 × 10 <sup>-10</sup>
α'-LiZr <sub>2</sub> P <sub>3</sub> O <sub>12</sub>	5.0 × 10 <sup>-8</sup>

- We adopt a binary classification strategy with a 10<sup>-4</sup> S/cm boundary, motivated by engineering requirements
- Training set includes 11 “good” conductors, 29 “bad” conductors



AD Sendek, Q Yang, ED Cubuk, KAN Duerloo, Y Cui, EJ Reed. *Ener. & Environ. Sci.*, doi:10.1039/C6EE02697D (2017)

We employ logistic regression (two-class classifier)

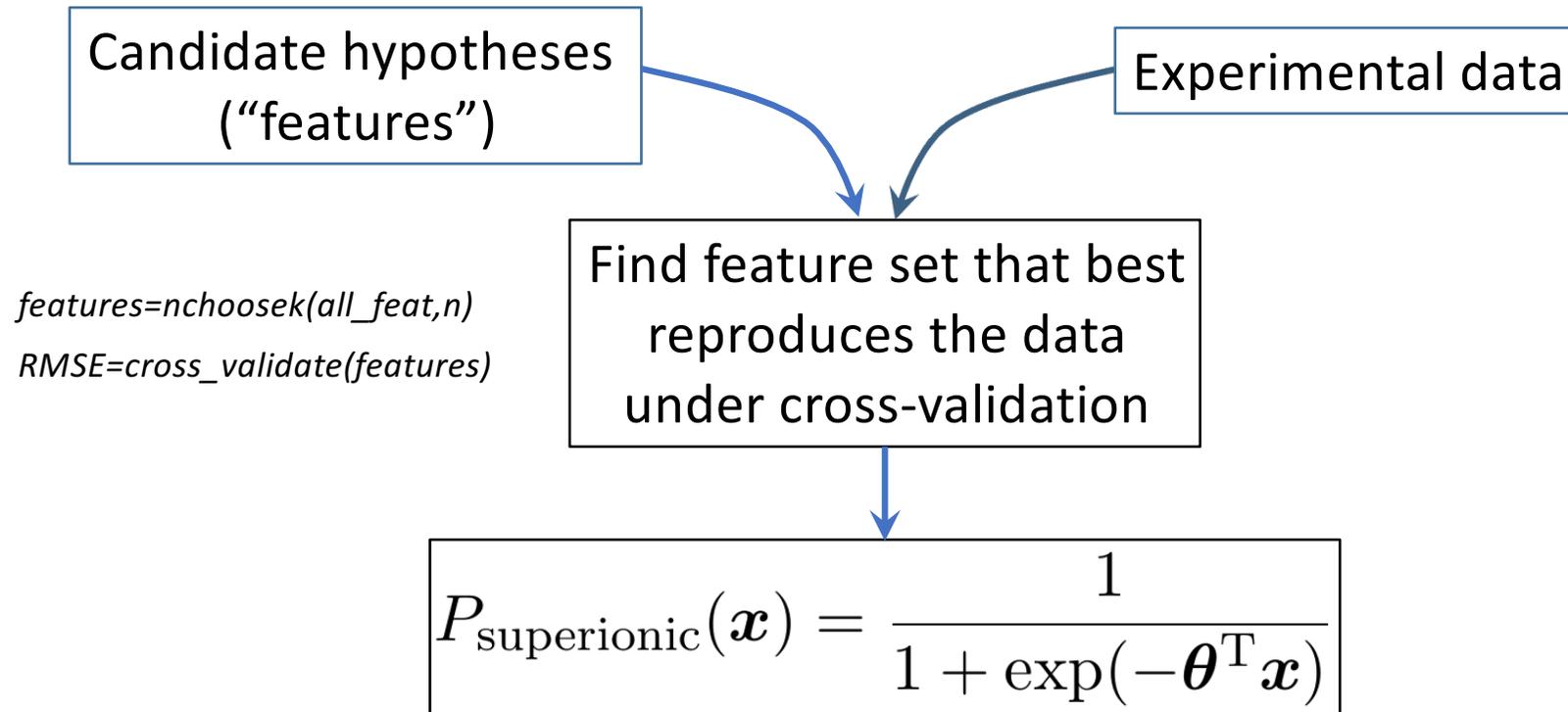
Assuming a logistic form, we search for the maximally predictive set of features:

$$P_{\text{superionic}}(\mathbf{x}) = \frac{1}{1 + \exp(-\boldsymbol{\theta}^T \mathbf{x})}$$

$$\boldsymbol{\theta}^T \mathbf{x} = ?$$

(Looping through data arrays = MATLAB's wheelhouse!)

We employ logistic regression (two-class classifier)



## Machine learning finds the best Li conduction model for this data set

$$P_{\text{superionic}}(\mathbf{x}) = \frac{1}{1 + \exp(-\boldsymbol{\theta}^T \mathbf{x})}$$

$$\boldsymbol{\theta}^T \mathbf{x} = 0.817LLB - 1.323SBI - 1.028AFC + 2.509LASD - 1.619LLSD - 1.944$$

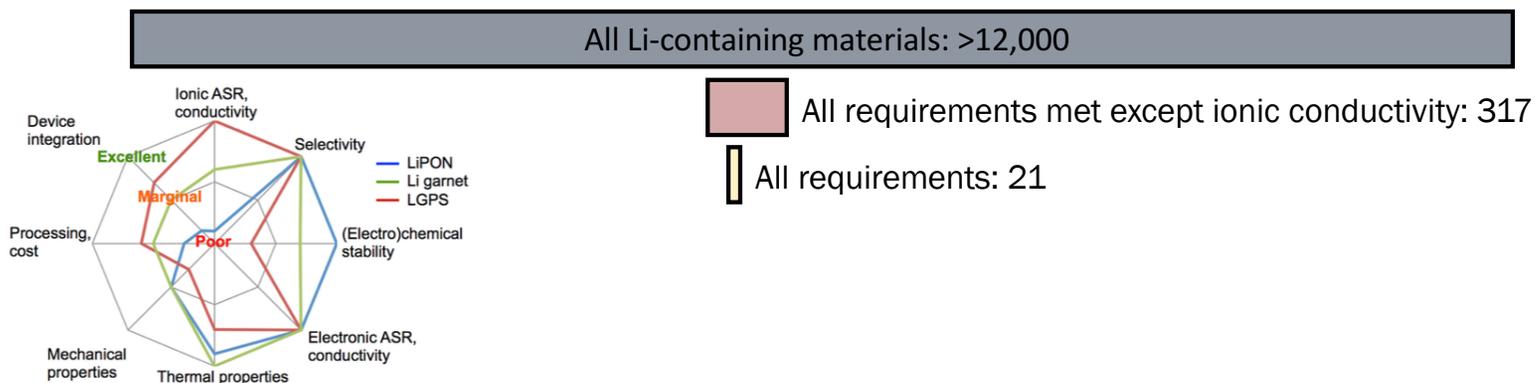
Lithium-lithium bond number      Sublattice bond ionicity      Anion framework coordination      Li-anion separation distance      Li-Li separation distance

>1 week with DFT  $\longrightarrow$  < 1 s with ML

# We perform the first holistic structure screening of all >12,000 candidates

Ionic conductivity is not all that matters! We also screen on:

- High stability against oxidation ← Gibbs free energy
- High stability against reduction ← Presence of transition metals
- Low electronic conductivity ← Band gap
- High phase stability ← Convex hull
- Low cost ← Cost of raw elements involved
- High earth abundance ← Abundance of elements in Earth's crust



AD Sendek, Q Yang, ED Cubuk, KAN Duerloo, Y Cui, EJ Reed. *Ener. & Environ. Sci.*, doi:10.1039/C6EE02697D (2017)

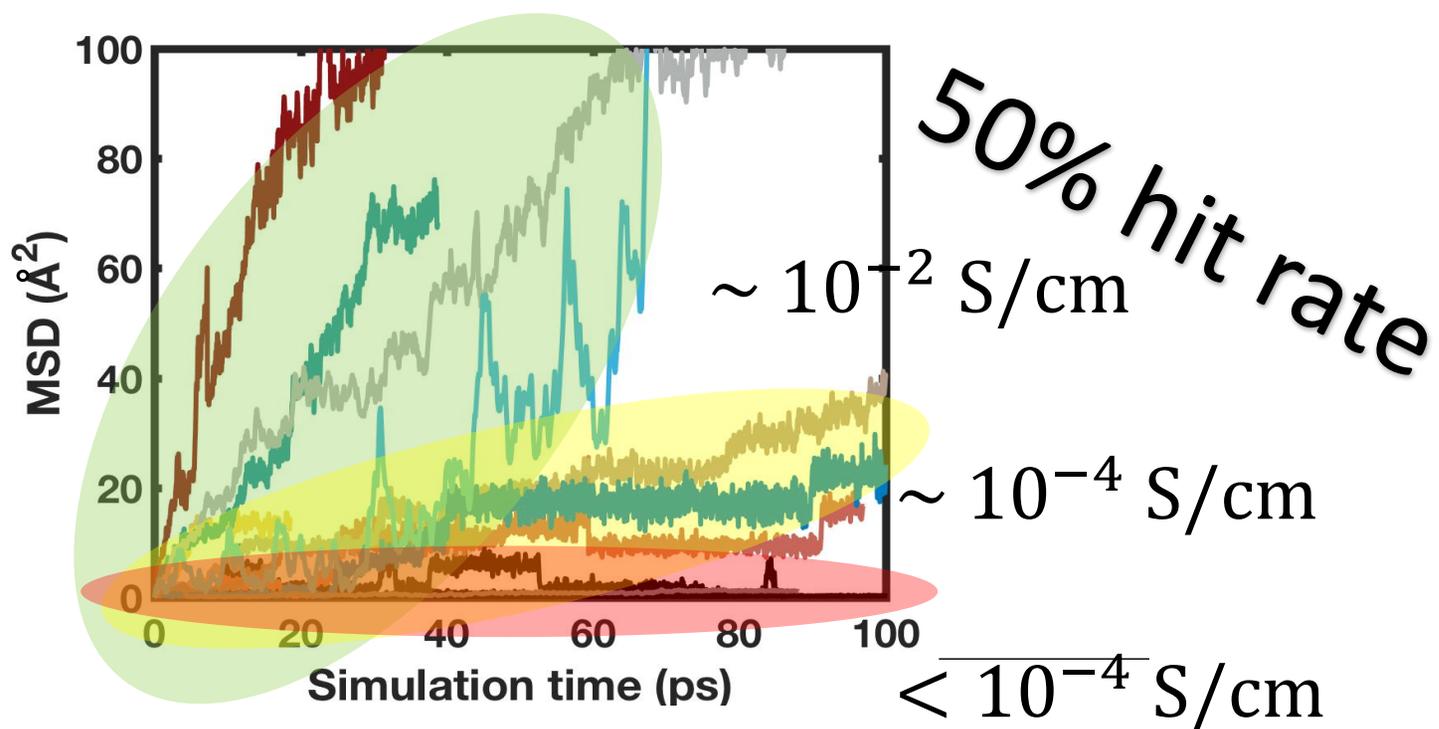
We publish 21 promising new candidates (without validation!)

MPID	Chemical formula	$P_{LR}$	$d$	$\epsilon$	$A$	$E_{gap}$	$\tilde{V}_{ox}$	USD/m <sup>2</sup> (10 $\mu$ m thick)	$I_A$	Related study
mp-554076	BaLiBS <sub>3</sub>	0.589	1.049	0.048	1	2.153	9.697	23	38	
mp-532413	Li <sub>5</sub> B <sub>7</sub> S <sub>13</sub>	0.897	1.228	0.024	1	3.553	5.454	42	38	95
mp-569782 <sup>a</sup>	Sr <sub>2</sub> LiCBr <sub>3</sub> N <sub>2</sub>	1.000	6.852	0.000	0	3.973	13.968	16	45	
mp-558219	SrLi(BS <sub>2</sub> ) <sub>3</sub>	0.518	1.556	0.114	1	2.91	13.964	38	38	
mp-15797	LiErSe <sub>2</sub>	0.543	1.505	0.056	1	1.615	6.778	170	67	
mp-29410	Li <sub>2</sub> B <sub>2</sub> S <sub>5</sub>	0.994	1.855	0.003	1	2.538	4.895	29	38	95
mp-676361	Li <sub>3</sub> ErCl <sub>6</sub>	0.655	0.974	0.042	1	5.211	7.794	70	44	96 and 97
mp-643069 <sup>a</sup>	Li <sub>2</sub> HfO	0.652	2.081	0.079	0	4.319	4.054	2.40	60	
mp-19896	Li <sub>2</sub> GePbS <sub>4</sub>	0.604	1.063	0.090	1	2.265	4.591	13	54	90
mp-7744 <sup>a</sup>	LiSO <sub>3</sub> F	1.000	4.097	0.000	0	5.792	13.446	10	34	
mp-22905 <sup>b</sup>	LiCl	0.837	1.381	0.031	1	6.25	4.214	0.94	34	98
mp-34477	LiSmS <sub>2</sub>	0.89	1.33	0.028	1	1.921	8.536	6.50	40	
mp-676109	Li <sub>3</sub> InCl <sub>6</sub>	0.656	1.013	0.058	1	3.373	6.215	5.50	63	96 and 97
mp-559238	CsLi <sub>2</sub> BS <sub>3</sub>	0.812	1.642	0.055	1	3.094	4.798	160	49	
mp-866665 <sup>a</sup>	LiMgB <sub>3</sub> (H <sub>9</sub> N) <sub>2</sub>	1.000	5.149	0.000	0	6.511	11.222	30	38	
mp-8751	RbLiS	0.775	1.279	0.051	1	2.745	4.22	240	34	
mp-15789	LiDyS <sub>2</sub>	0.901	1.339	0.025	1	1.935	8.736	9.20	39	
mp-15790	LiHoS <sub>2</sub>	0.899	1.327	0.025	1	1.965	8.749	300	55	
mp-15791	LiErS <sub>2</sub>	0.899	1.319	0.025	1	2.008	8.761	190	44	
mp-561095 <sup>a</sup>	LiHo <sub>3</sub> Ge <sub>2</sub> (O <sub>4</sub> F) <sub>2</sub>	0.984	3.247	0.009	0	4.163	53.18	370	55	
mp-8430	KLiS	0.76	1.243	0.052	1	3.057	4.348	14	34	

...and hope we're right

How well does the model do?

## We discover ten new solids that are superionic conductors



# We discover ten new solids that are superionic conductors

## ML-guided simulations

Name	$P_{\text{superionic}}$
BaLiBS <sub>3</sub>	58.9%
Li <sub>5</sub> B <sub>7</sub> S <sub>13</sub>	89.7%
Sr <sub>2</sub> LiCBr <sub>3</sub> N <sub>2</sub>	100%
SrLi(BS <sub>2</sub> ) <sub>3</sub>	51.8%
LiErSe <sub>2</sub>	54.3%
Li <sub>2</sub> B <sub>2</sub> S <sub>5</sub>	99.4%
Li <sub>3</sub> ErCl <sub>6</sub>	65.5%
Li <sub>2</sub> HfO	65.2%
Li <sub>2</sub> GePbS <sub>4</sub>	60.4%
LiSO <sub>3</sub> F	100%
LiCl	83.7%
LiSmS <sub>2</sub>	89.0%
Li <sub>3</sub> InCl <sub>6</sub>	65.6%
CsLi <sub>2</sub> BS <sub>3</sub>	81.2%
LiMgB <sub>3</sub> (H <sub>9</sub> N) <sub>2</sub>	100%
RbLiS	77.5%
LiDyS <sub>2</sub>	90.1%
LiHoS <sub>2</sub>	89.9%
LiErS <sub>2</sub>	89.9%
LiHo <sub>3</sub> Ge <sub>2</sub> (O <sub>4</sub> F) <sub>2</sub>	98.4%
KLiS	76%

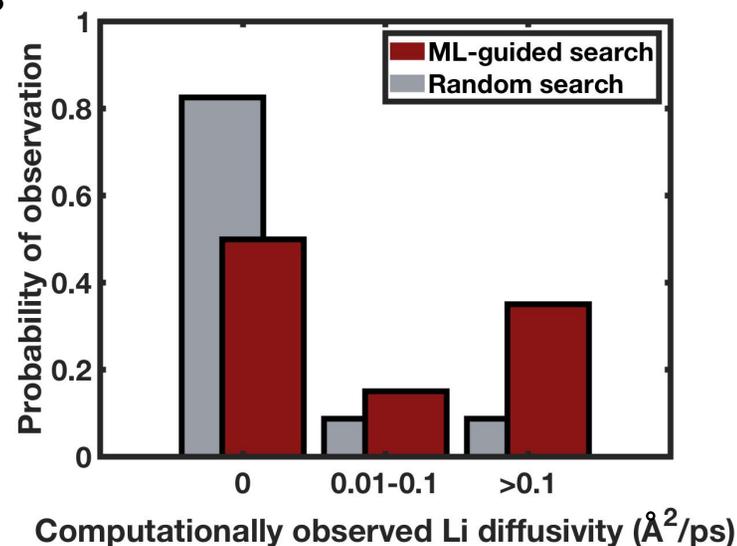
Model success  
rate: ~35%  
(melting cases excluded)

Random success  
rate: ~10%  
(melting cases excluded)

## Random simulations

Name	$P_{\text{superionic}}$
LiLa <sub>2</sub> SbO <sub>6</sub>	0
Li <sub>6</sub> UO <sub>6</sub>	7.70%
Li <sub>4</sub> H <sub>3</sub> BrO <sub>3</sub>	43%
LiInF <sub>4</sub>	0.60%
LiBiF <sub>4</sub>	0.20%
CsLi <sub>2</sub> (HO) <sub>3</sub>	3.50%
Li <sub>6</sub> Ho(BO <sub>3</sub> ) <sub>3</sub>	10.10%
RbLiB <sub>4</sub> O <sub>7</sub>	4%
LiU <sub>4</sub> P <sub>3</sub> O <sub>20</sub>	0.10%
Li <sub>4</sub> Be <sub>3</sub> As <sub>3</sub> ClO <sub>12</sub>	0.30%
Li <sub>6</sub> TeO <sub>6</sub>	6.40%

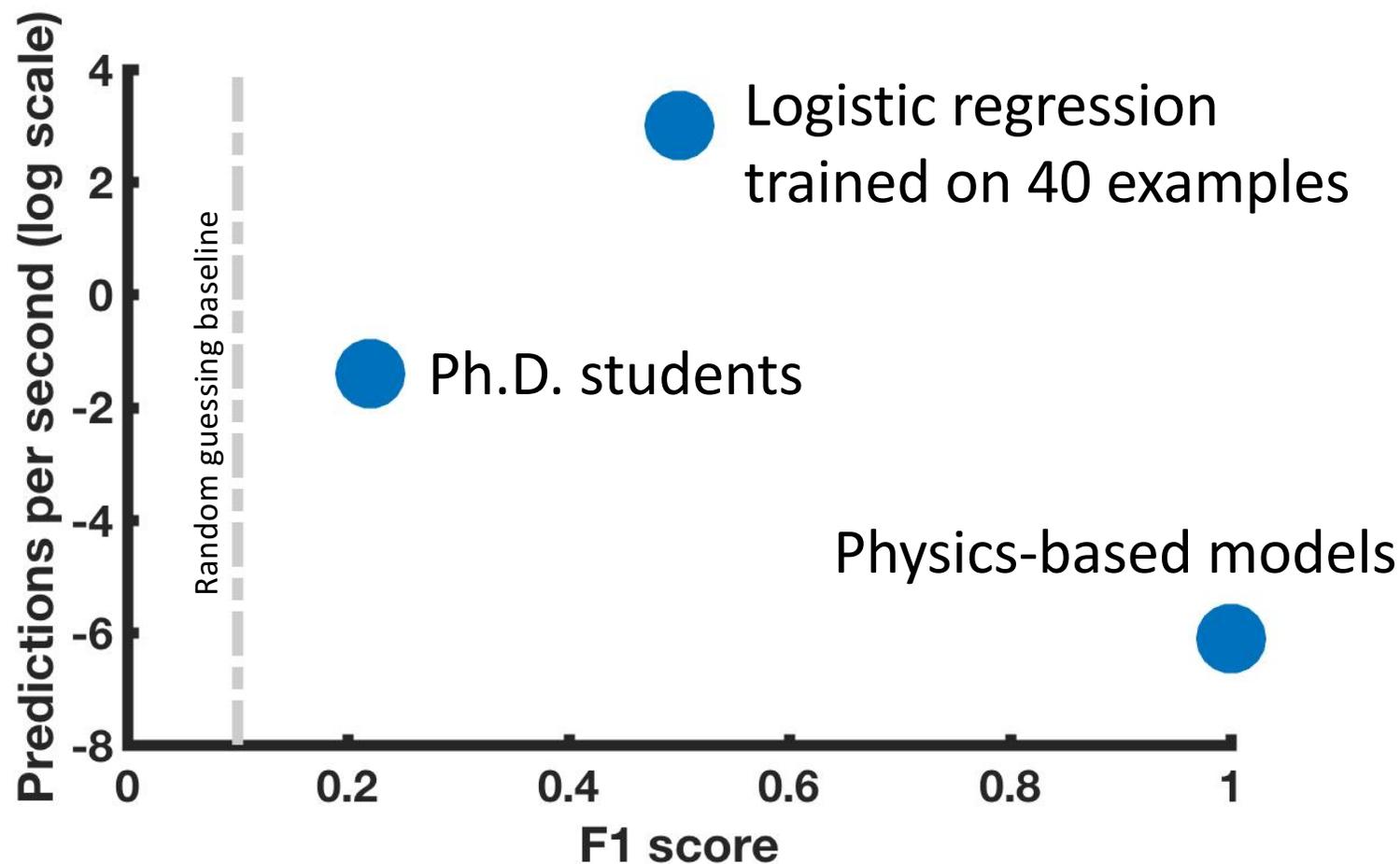
⋮ ⋮



Learning from only 40 data points = 3x improvement over guesswork

+ exciting, new, potentially record-setting material!

Our algorithm outperforms humans in accuracy and speed



AD Sendek, ED Cubuk, G Cheon, Y Cui, EJ Reed. In preparation. (2017)

## We leverage machine learning to identify the most promising solid electrolytes $10^6$ x faster than state-of-the-art

- Our model is  $\sim 3$ x better than random guessing
- Achieved by collecting most of the available data and proposed wisdom over past four decades
- We perform the first holistic screening of all known Li containing solids and discover several new superionic conducting structures
- Exciting new materials for further development!

**Screening:** AD Sendek, Q Yang, ED Cubuk, KAN Duerloo, Y Cui, EJ Reed. *Ener. & Environ. Sci.*, doi:10.1039/C6EE02697D (2017).  
**DFT MD:** AD Sendek, ED Cubuk, Y Cui, EJ Reed. In preparation.

**40 data points = 3x improvement**

**Imagine what we can do with 100, 500, 1000?**

# The value we get from MATLAB

## **Quick prototyping**

- MATLAB is easy and intuitive to use; get a prototype out the door faster

## **Text file processing**

- Very easy to scan through directories and read in files

## **Thorough, up-to-date documentation**

- Can always get answers quickly

## **Figure generation**

- All our figures are made in MATLAB; it is easy and the plots are clean



Thank you!

Austin Sendek

Email: [asendek@stanford.edu](mailto:asendek@stanford.edu)

Web: [stanford.edu/~asendek](http://stanford.edu/~asendek)

Prof. Evan Reed: [evanreed@stanford.edu](mailto:evanreed@stanford.edu)