Supplemental Material for: “Automated discovery of a robust interatomic potential for aluminum”

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Supplementary Note 1 DFT Calculations

We used the Quantum Espresso [1, 2] software package, using a 3 electron PBE ultrasoft pseudopotential (Al.pbe-n-rrkjus_psl.0.1.UPF). All calculations we performed using a kinetic energy cutoff of 20Ry and a $3 \times 3 \times 3$ k-point grid. In retrospect, a larger basis would have been more appropriate for the linear dimensions of our supercells, ranging from 10.5 Å to 17.0 Å (units cells contain up to 250 atoms). We used the Marzari-Vanderbilt [3] method of thermal electron smearing with an electronic temperature of 0.06 Ry. The SCF iterations were converged to within $10^{-8}$ Ry. Each Quantum Espresso calculation was parallelized over four Nvidia V100 GPUs; the typical wall clock time for a QE calculation with 124 atoms was 8 minutes.

Supplementary Note 2 Active Learning Details

To bootstrap the active learning procedure, we generate an initial data set of 401 DFT calculations, performed on randomized atomic configurations. Each such configuration contains between 55 to 249 Al atoms, randomly placed in an orthorhombic supercell, subject to a non-overlapping constraint, as described in the main text. Active learning proceeds in iterations. At each iteration, ANI neural networks are trained on all available DFT data, and used to carry out molecular dynamics (MD) simulations for the purpose of sampling. We use an MD step size of 1 fs. MD initial conditions are randomized. To diversify the sampling, each MD run is driven according to a randomized heating and cooling schedule as described in the main text. An uncertainty quantification technique is utilized to determine whether the ANI-Al model (under construction) is confident in its predictions for the current configuration of atoms. To estimate model uncertainty, we employ the ensemble disagreement technique used in previous work [4]. Specifically, we look at the sample

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variance $\text{Var}[\hat{E}]$ of the energy predictions $\hat{E}$ over the eight neural nets that comprise a single (enssembled) ANI-Al model. We also look at the ensemble variance for the force predictions, $\text{Var}[\hat{f}]$, averaged over all $N$ atoms in the unit cell. If, during active learning, the ensemble variance for energy or force prediction exceeds a threshold ($E_{\text{thresh}} = 0.001\sqrt{N}\text{eV}$ or $f_{\text{thresh}} = 0.002\text{eV/Å}$, respectively) then a new DFT calculation is queued to help the ML model to better understand that atomic configuration. If an uncertainty threshold is exceeded, we halt MD simulation and queue a DFT calculation to acquire ground-truth training data for the current MD configuration. If an MD simulation reaches 8 hours of wall-clock time (about 200ps to 500ps of simulated time) without reaching the uncertainty threshold, it is respawned. Batches of MD simulations are carried out with multiple ANI-Al models simultaneously. DFT calculations are continuously performed on the queued configurations. If 2 hours pass since the last ANI-Al model was trained and more than 25 new data points had been generated, a new ANI-Al model is trained. In this way, full cycles of ensemble training, MD simulations for sampling, and DFT data generation are carried out simultaneously to build the training data set. Our final training data set contains 6,352 DFT calculations. Each DFT calculation provides considerable data: a supercell contains between 55 and 249 Al atoms, and DFT calculates forces for each atom.

Figure 7 below illustrates the workflow for the MD simulations used during active learning.

**Supplementary Note 3 ANI-Al Model Hyper-parameters**

The ANI neural networks used in this work were implemented in the NeuroChem C++/CUDA software package. A batch size of 128 was used while training the ANI-Al model. A weight of 1.0 was used on the energy loss term of the loss function, while a weight of 0.01 was used on the force training loss term. Learning rate annealing was used during training, starting at a learning rate of 0.001 and converging at a learning rate of 0.00001. The learning rate annealing algorithm is described in previous work [5]. The ADAM [6] update algorithm is used during training. The network architecture is provided in Table 1.
All ANI-Al model symmetry function parameters are provided below:

Radial Cutoff (Radial): 7.0
Radial Cutoff (Angular): 5.0
Radial Eta: [43.9]
Radial Shift: [1.2500000, 1.4296875, 1.6093750e, 1.7890625, 1.9687500e,
2.1484375, 2.3281250, 2.5078125, 2.6875000, 2.8671875, 3.0468750,
3.2265625, 3.4062500, 3.5859375, 3.7656250, 3.9453125, 4.1250000,
4.3046875, 4.4843750, 4.6640625, 4.8437500, 5.0234375, 5.2031250,
5.3828125, 5.5625000, 5.7421875, 5.9218750, 6.1015625, 6.2812500,
6.4609375, 6.6406250, 6.8203125]
Angular Zeta: [69.4]
Angular Angular Shift: [0.19634954, 0.58904862, 0.98174770, 1.3744468e,
2.1598449, 2.5525440, 2.9452431]
Angular Eta: [6.5]
Angular Radial Shift: [1.2500000, 1.7187500, 2.1875000, 2.6562500]

Supplementary Note 4 Nearsightedness principle and the interaction cutoff distance

The ANI-Al predicted total system energy \( \hat{E} = \sum_i \hat{E}_i \) is a sum over local contributions centered on each atom \( i \). For the hyperparameters listed above, each local energy \( \hat{E}_i \) incorporates information about neighboring atoms \( j \) if the pairwise distance \( r_{ij} = |r_i - r_j| \) is less than 7 Å. Furthermore, \( \hat{E}_i \) can incorporate angular information \( r_{ij} \cdot r_{ik} \) involving neighboring atoms \( j \) and \( k \) if both are within a smaller cutoff distance: \( r_{ij} < 5 \) Å and \( r_{ik} < 5 \) Å. Using \( i \) as an intermediary, the total ANI-Al energy can strongly couple atoms \( j \) and \( k \) separated by a distance of up to 10 Å, even though the “cutoff distance” is just 7 Å.

The ANI-Al predicted force on atom \( j \) is \( f_j = -d\hat{E}/dr_j \). Note that \( f_j \) involves \( -d\hat{E}_i/dr_j \) if \( r_{ij} < 7 \) Å. In turn, \( \hat{E}_i \) can depend on atom \( k \) if \( r_{ik} < 7 \) Å. Considering a linear geometry in which atoms \( j, i, \) and \( k \) are separated by 7 Å intervals, one observes that \( f_j \) may in principle be affected by an atom \( k \) for pairwise distances of up to \( r_{jk} < 14 \) Å.

Supplementary Note 5 Training to forces

We employed the loss function

\[
L = \left( \hat{E} - E \right)^2 + \ell_0^2 \sum_{j=1}^{N} \left( \hat{f}_j - f_j \right)^2,
\]

summed over systems in the dataset.

Stochastic gradient descent training requires calculation of all components \( \partial L/\partial W_i \) of the loss gradient. Typically, there are order \( 10^5 \) model parameters \( W_i \).

Frameworks such as TensorFlow or PyTorch support iterated backpropagation, thereby enabling efficient calculation of the full gradient \( \partial L/\partial W_i \) at a cost comparable to calculating \( L \) itself [7, 8, 9].

For the present work, we used the C++ Neurochem implementation of ANI [10], for which iterated backpropagation would be challenging to implement. Instead, we use a recently developed
scheme to train to force data [11]. The force part of the loss gradient \( \partial L / \partial W_i \) can be cast as a directional derivative of \( \partial \hat{E}_i[r] / \partial W_i \), in which each atomic position \( r_j \) is varied in the direction of the observed force prediction error. Using central differences, the loss gradient may be approximated as

\[
\frac{\partial L}{\partial W_i} = 2 \left( \hat{E} - E \right) \frac{\partial \hat{E}}{\partial W_i} - \frac{\eta}{\partial \hat{E}} \left( \frac{\partial \hat{E}[r^+]}{\partial W_i} - \frac{\partial \hat{E}[r^-]}{\partial W_i} \right)^2,
\]

where \( r_j^\pm = r_j \pm \eta (\hat{f}_j - f_j) \) represents a carefully selected perturbation to the position of atom \( j \) (for all \( j \) simultaneously). Crucially, \( r_j^\pm \) is treated as fixed with respect to variations in model parameters \( W_i \). This central difference approximation would be exact in the limit that \( \eta \to 0 \).

Guidance on selecting \( \eta \) at fixed floating point precision is provided in Ref. [11]. This scheme is highly efficient because, for a given atomic configuration, the full energy gradient \( \partial \hat{E} / \partial W_i \), for all model parameters \( W_i \), can be efficiently calculated using straightforward backpropagation (i.e., using the same techniques as needed to implement energy-only training). In practice, we find that the cost to calculate all \( \partial L / \partial W_i \) is of the same order as the cost to calculate all forces.

**Supplementary Note 6 t-SNE Embeddings**

For each atom in the active learning generated data set we obtain a chemical environment descriptor, which is the first hidden layer of an ANI-Al model. These chemical environment descriptors are embedded into a 2D space using the OpenTSNE python package [12]. The t-SNE algorithm is initialized with principal component analysis (PCA). The cosine distance metric is used with a perplexity of 100.

**Supplementary Note 7 MD Simulation Details**

**Melting temperatures via solid-liquid coexistence.** The melt-curve was determined using solid-liquid coexistence simulations [13, 14, 15] performed in the NPT ensemble. Independent NPT simulations were performed to determine equilibrium densities of solid/liquid Al determined at the relevant (P,T) conditions. The starting configurations for solid-liquid coexistence simulations, containing 16000 atoms, were created using these equilibrium densities. The simulation cells were equilibrated in NVT ensemble for 100 ps, at a temperature close to the expected melting point. At the end of the equilibration, the simulation cell consisted of liquid Al in contact with the (100) crystallographic face of Al, with roughly equal number of atoms in the solid and liquid phases. Following this, independent NPT simulations of solid-liquid coexistence were performed at a set of temperatures spanning the melting temperature at a given pressure. The time evolution of the simulation-cell volume was used to determine the upper and lower bounds of the melting temperature, i.e., an increase/decrease in the simulation cell volume with time implied that the system was above/below the melting temperature. All the melting-points presented in Figure 6 of the main article are the upper bounds determined as explained. Lower bounds are at the most 25 K lower than the reported upper bounds.

We note that a fcc-bcc phase transition was predicted for the Mendelev-EAM potential [16] at P > 20 GPa. Therefore, we calculated the melt-curve for this potential only up to 15 GPa.

**Phase Transition Dynamics.** For this simulation, we first slowly heat the system from 300 K to 1500 K, passing through the melt point of 933 K at atmospheric pressure. Next we slowly cool the system back to 300 K. The entire heating-cooling process runs for 750 ps. As with our previous MD simulations, we use a Langevin thermostat with a friction coefficient of 0.02 fs\(^{-1}\), and we apply
an MD timestep of 0.5 fs. The simulation is performed in a periodic box containing 108 atoms initialized in an FCC lattice (that is, \(3 \times 3 \times 3\) cubic unit cells). The density of the simulation is fixed by selecting a volume of \((12.138\,\text{Å})^3\). At this density, the simulated pressure should be about 1 bar at the coexistence temperature.

**Variable temperature and density liquid phase simulations.** We iterated through five temperatures, 1000 K, 1200 K, 1400 K, 1600 K, and 1800 K. At each temperature, we used ANI-Al to drive MD simulations of 108 atoms at five volumes, \(0.850V_0\), \(0.925V_0\), \(1.00V_0\), \(1.075V_0\), and \(1.15V_0\), where \(V_0\) is the mean volume at zero pressure. Each simulation was carried out for 60 ps. We sampled 10 snapshots from each of the \(5 \times 5\) MD trajectories. Figure S5 shows the accuracy of the ANI-Al model on each trajectory.

### Supplementary Table 1: ANI-Al active learning trained model architecture.

| Layer ID | Nodes | Activation |
|----------|-------|------------|
| 1        | 96    | CELU       |
| 2        | 96    | CELU       |
| 3        | 64    | CELU       |
| 4        | 1     | Linear     |

### Supplementary Table 2: Holdout test set performance for ANI and HIP-NN models on the Al data set. These are single model, not ensemble errors.

| Property                | ANI   | HIP-NN |
|-------------------------|-------|--------|
| Energy RMSE (meV/atom)  | 2.64  | 3.45   |
| Force RMSE (eV/Å)       | 0.067 | 0.130  |
Supplementary Table 3: Various calculated/experimental properties for FCC aluminum: lattice constant $a$, bulk modulus $B$, elastic constants $C$, energy of formation for vacancy ($E_{\text{vac}}^f$) and (100) dumbbell interstitial ($E_{\text{inter}}^f$) defects, surface energy $E_{\text{surf}}$ (experimental data is averaged over orientations), surface relaxation the between the two outermost layers ($d_{12}$) and just below that ($d_{23}$), unstable stacking fault energy $E_{\text{usf}}$, stacking fault energy $E_{\text{sf}}$, unstable twinning fault energy $E_{\text{utf}}$, and melting temperature at atmospheric pressure $T_{\text{melt}}$.

| Property               | Li04   | Me08   | Mi99   | Zo03   | Sh10   | Wi09   | Pa15   | ANI    | DFT    | Expt.   |
|------------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---------|
| $a$ (Å)                | 4.032  | 4.045  | 4.050  | 4.050  | 4.018  | 4.025  | 4.054  | 4.042  | 4.046 ± 0.004 [17] |
| $B$ (GPa)              | 81.6   | 75.1   | 79.0   | 79.0   | 82.6   | 79.1   | 78.9   | 77.3   | 76.7   | 79.0 [18] |
| $C_{11}$ (GPa)         | 119    | 106    | 114    | 117    | 119    | 114    | 114    | 117    | 106    | 114 [18] |
| $C_{12}$ (GPa)         | 62.9   | 59.5   | 61.6   | 60.1   | 64.1   | 61.7   | 61.6   | 57.2   | 62.3   | 61.9 [18] |
| $C_{44}$ (GPa)         | 32.9   | 30.7   | 31.6   | 31.7   | 30.8   | 31.3   | 45.4   | 30.4   | 31.6   | 31.6 [18] |
| $E_{\text{vac}}^f$ (meV) | 683    | 659    | 676    | 709    | 673    | 664    | 671    | 663    | 618    | 680 [19], 660 ± 20 [20] |
| $E_{\text{inter}}^f$ (eV) | 2.72   | 2.34   | 2.60   | 2.21   | 2.29   | 2.61   | 3.03   | 2.49   | 2.85   | -       |
| $E_{100}^\text{surf}$ (mJ m$^{-2}$) | 1002   | 496    | 944    | 607    | 842    | 912    | 977    | 990    | 929    | 902 ± 78 [21], 980 [22], |
| $E_{110}^\text{surf}$ (mJ m$^{-2}$) | 1107   | 581    | 1006   | 793    | 916    | 1047   | 1123   | 1015   | 997    | 902 ± 78 [21], 980 [22], |
| $E_{111}^\text{surf}$ (mJ m$^{-2}$) | 912    | 428    | 871    | 601    | 813    | 876    | 718    | 855    | 827    | 902 ± 78 [21], 980 [22], |
| $d_{12}^{100}$ (%)     | -2.22  | -4.15  | -0.57  | -5.89  | -1.93  | -0.41  | 10.54  | -1.02  | 1.21   | 2.0 ± 0.8 [23] |
| $d_{23}^{100}$ (%)     | -1.69  | -0.84  | -0.92  | -4.15  | -1.69  | -0.73  | 2.54   | 0.69   | 0.2    | 1.2 ± 0.7 [23] |
| $d_{12}^{110}$ (%)     | -4.09  | -7.66  | -1.53  | -9.10  | -3.20  | -3.01  | 8.20   | -10.63 | -8.11  | -8.5 ± 1.0 [24] |
| $d_{23}^{110}$ (%)     | 1.85   | -0.34  | 1.53   | -1.85  | 0.41   | 1.17   | 6.33   | 6.91   | 4.34   | 5.5 ± 1.0 [24] |
| $d_{12}^{111}$ (%)     | 0.36   | -3.66  | 0.02   | -4.49  | -1.15  | -0.30  | 7.90   | 0.7    | 1.27   | 0.9 ± 0.7 [25] |
| $d_{23}^{111}$ (%)     | -0.21  | -0.12  | -0.30  | -1.71  | -0.91  | -0.05  | 2.21   | 0.03   | 0.22   | -       |
| $E_{\text{usf}}$ (mJ m$^{-2}$) | 163    | 220    | 168    | 150    | 143    | 180    | 307    | 181    | 179    | -       |
| $E_{\text{sf}}$ (mJ m$^{-2}$) | 131    | 127    | 146    | 114    | 117    | 141    | 187    | 144    | 159    | 166 [22], 120 [26] |
| $E_{\text{utf}}$ (mJ m$^{-2}$) | 204    | 276    | 220    | 198    | 181    | 234    | 350    | 237    | 254    | -       |
| $T_{\text{melt}}$ (K)  | 930    | 925    | 1050   | 875    | 740    | 850    | 950    | 925    | -      | 933 [27] |
Supplementary Table 4: Comparison of FCC property predictions for three ML potentials of aluminum: Physically Informed Neural Network (PINN) [28], DeepPot (DP) [29], and the present work (ANI-Al). Percent errors are given with respect to the reference DFT calculations original reported for each model.

|                | PINN DFT | PINN DFT | DP DFT | ANI-Al DFT | ANI-Al DFT | PINN Error | DP Error | ANI-Al Error |
|----------------|----------|----------|--------|------------|------------|------------|----------|--------------|
| \(a\) (Å)      | 4.039    | 4.039    | 4.042  | 4.054      | 0.01%      | 0.06%      | 0.30%    |
| \(B\) (GPa)    | 83       | 79       | 78     | 80.1       | 76.7       | -4.82%     | 2.69%    | 0.78%        |
| \(C_{11}\) (GPa) | 104     | 117      | 111    | 120.9      | 106        | 117        | 12.50%   | 8.92%        |
| \(C_{12}\) (GPa) | 73      | 60       | 61.4   | 59.6       | 62.3       | 57.2       | -17.81%  | -2.93%       |
| \(C_{14}\) (GPa) | 32      | 32       | 36.8   | 40.4       | 31.6       | 30.41      | 0.00%    | 9.78%        |
| \(E_{surf}^{100}\) (mJ m\(^{-2}\)) | 920    | 899      | 920    | 950        | 929        | 990        | -2.28%   | 3.26%        |
| \(E_{surf}^{110}\) (mJ m\(^{-2}\)) | 980    | 952      | 980    | 999        | 1015       | 1015       | -2.86%   | 1.02%        |
| \(E_{surf}^{111}\) (mJ m\(^{-2}\)) | 800    | 819      | 800    | 820        | 827        | 855        | 2.38%    | 2.50%        |
| \(E_{vac}^{I}\) (meV) | 665    | 678      | 670    | 790        | 618        | 663        | 1.95%    | 17.91%       |
| \(E_{inter}^{I}\) (eV) | 2.607  | 2.246    | -      | -          | 2.85       | 2.49       | -13.85%  | -12.63%      |
| \(E_{uf}\) (mJ m\(^{-2}\)) | 158    | 121      | 142    | 132        | 159        | 144        | -23.42%  | -7.04%       |
| \(E_{usf}\) (mJ m\(^{-2}\)) | 175    | 132      | -      | -          | 179        | 181        | -24.57%  | -1.12%       |

Supplementary Table 5: Theoretically predicted crystal energies, relative to the ground state. Crystals included are: diamond cubic, simple cubic, A5, L1\(_2\) (face-centered cubic with one vacant site), body-centered cubic, A15, hexagonal close packed, 9R, double hexagonal close packed, and face-centered cubic. Units are eV/atom.

| Crystal       | Li04 | Me08 | Si99 | Zo03 | Sh10 | Wi09 | Pa15 | ANI-Al | DFT  |
|---------------|------|------|------|------|------|------|------|--------|------|
| DC            | 0.8879 | 0.8123 | 0.8909 | 0.8842 | 0.7006 | 1.0388 | 1.0633 | 0.6788 | 0.7333 |
| SC            | 0.3557 | 0.3124 | 0.3986 | 0.2966 | 0.2298 | 0.2208 | 0.39   | 0.386  | 0.3695 |
| A5 (\(\beta\)-Sn) | 0.2966 | 0.2664 | 0.3241 | 0.2182 | 0.1752 | 0.1906 | 0.2357 | 0.2592 | 0.2722 |
| L1\(_2\)      | 0.2621 | 0.2863 | 0.302  | 0.3292 | 0.2403 | 0.2738 | 0.0732 | 0.2381 | 0.204  |
| BCC           | 0.0842 | 0.1019 | 0.1055 | 0.0926 | 0.0998 | 0.1295 | 0.1587 | 0.1006 | 0.0925 |
| A15 (\(\beta\)-W) | 0.0103 | 0.0599 | 0.0131 | 0.0165 | 0.074  | 0.0834 | 0.1855 | 0.0781 | 0.0749 |
| HCP           | 0.0237 | 0.0276 | 0.0278 | 0.0216 | 0.0244 | 0.0301 | 0.0401 | 0.0312 | 0.0293 |
| 9R            | 0.0179 | 0.0186 | 0.0205 | 0.0161 | 0.0168 | 0.0204 | 0.0271 | 0.0211 | 0.0181 |
| DHCP          | 0.0152 | 0.0141 | 0.0161 | 0.0133 | 0.0128 | 0.0154 | 0.0204 | 0.0161 | 0.0104 |
| FCC           | 0     | 0     | 0     | 0     | 0     | 0     | 0     | 0      | 0     |
Supplementary Figure 1: Distribution of pressures in the training data set.
Supplementary Figure 2: Complete path errors for each method compared to reference DFT data. The methods are order from left to right by the lowest average error.
Supplementary Figure 3: Correlation plots of energy, forces, and pressures for liquid Al MD trajectories over a grid of atomic volumes (0.8Å³ to 1.2Å³) and temperatures (1000K to 2000K).
Supplementary Figure 4: Plots of liquid Al MD trajectories over a grid of atomic volumes (0.8Å³ to 1.2Å³) and temperatures (1000K to 2000K).
Supplementary Figure 5: Energy correlations. “FCC/Melt” refers to the dataset collected near equilibrium for these two phases. “Disorder” refers to the dataset collected automatically by the active learning procedure (the AL dataset of the main text), and includes highly out of equilibrium data.
Supplementary Figure 6: Force component correlations.
Supplementary Figure 7: Diagram of the active learning algorithm.
Supplementary Figure 8: Thermal expansion of ANI-Al compared to experiment and other EAM models.
Supplementary Figure 9: Extended cold curves for ANI-Al ensemble. At zero temperature ANI-Al predicts an FCC-to-HCP transition at 154.8 GPa, and an HCP-to-BCC transition at 392.7 GPa. The BCC crystal becomes lower energy than FCC above 261.5 GPa.

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