Supplementary Information

A Continuous Action Space Tree search for INverse desiGn (CASTING) Framework for Materials Discovery

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S. 1. Calculation of the unique structures sampled by MCTS, GA, and random sampling for C polymorphs:

To calculate the unique C structures sampled during a search of any sampling algorithm (e.g., MCTS, GA, random), we follow a two-step approach. We start by calculating order parameters (Q_2, Q_4, Q_6)[1] + coordination vectors of the sampled structures with a cutoff of 3A^o. We then use an unsupervised Agglomerative Clustering[2], with a "distance_threshold" of 1 and a "ward" linkage to cluster similar configurations in feature space. Thus, the number of clusters is the number of unique structures sampled and from each cluster, the configuration having the least energy value is chosen.

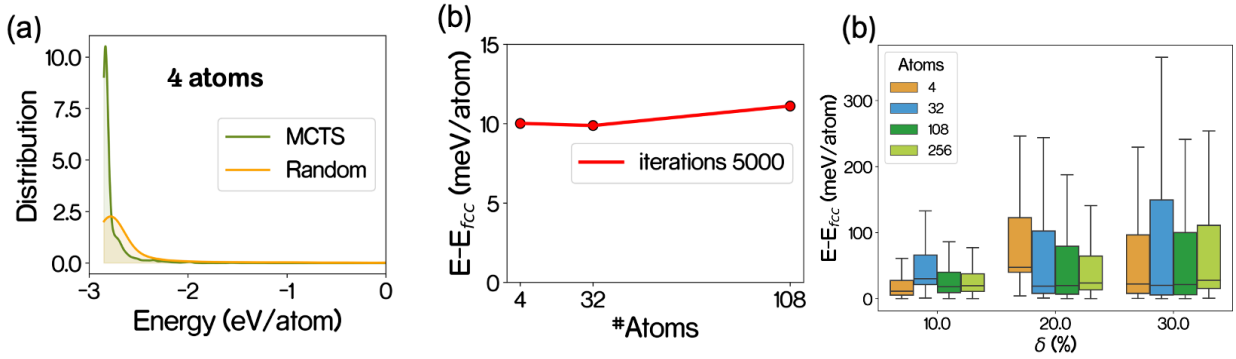

Fig. S1. *(a) Typical energy distribution of Ag FCC structure sampled by MCTS algorithm as compared to a purely random sampling approach for a system size of 4 atoms. (b) The effect of an increase in the number of dimensions on the best solution obtained for a bound increment (δ) of 30%, using a random sampling approach. (c) The overall energy distribution of all the sampled configurations for varying dimesionalities with different lattice parameter bounds (δ).*

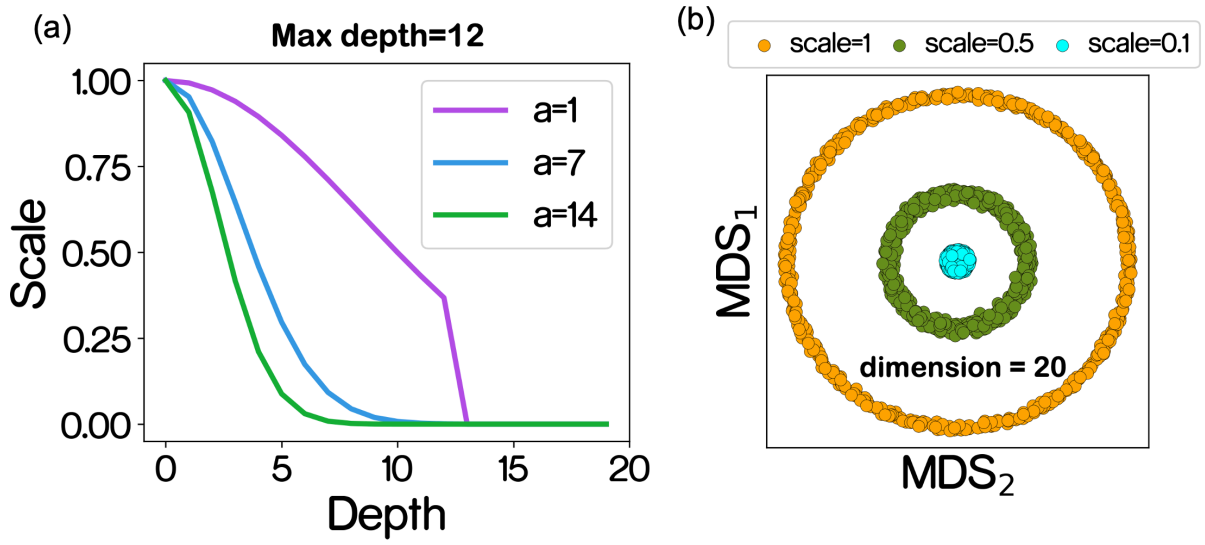

Fig S2. *(a) Depth scaling for different values of "a" hyperparameter. The scale reduces following Eq.2. (b) The MDS (Multi-Dimensional Scaling) plot of parameter vectors (20 dimensions) following a Hypersphere perturbation scheme for the mutation of lattice parameters and coordinates of the atoms. The points are sampled on the surface of the hypersphere, around the target point in search space. The perturbation window is scaled with depth by changing the scaling factor "a".*

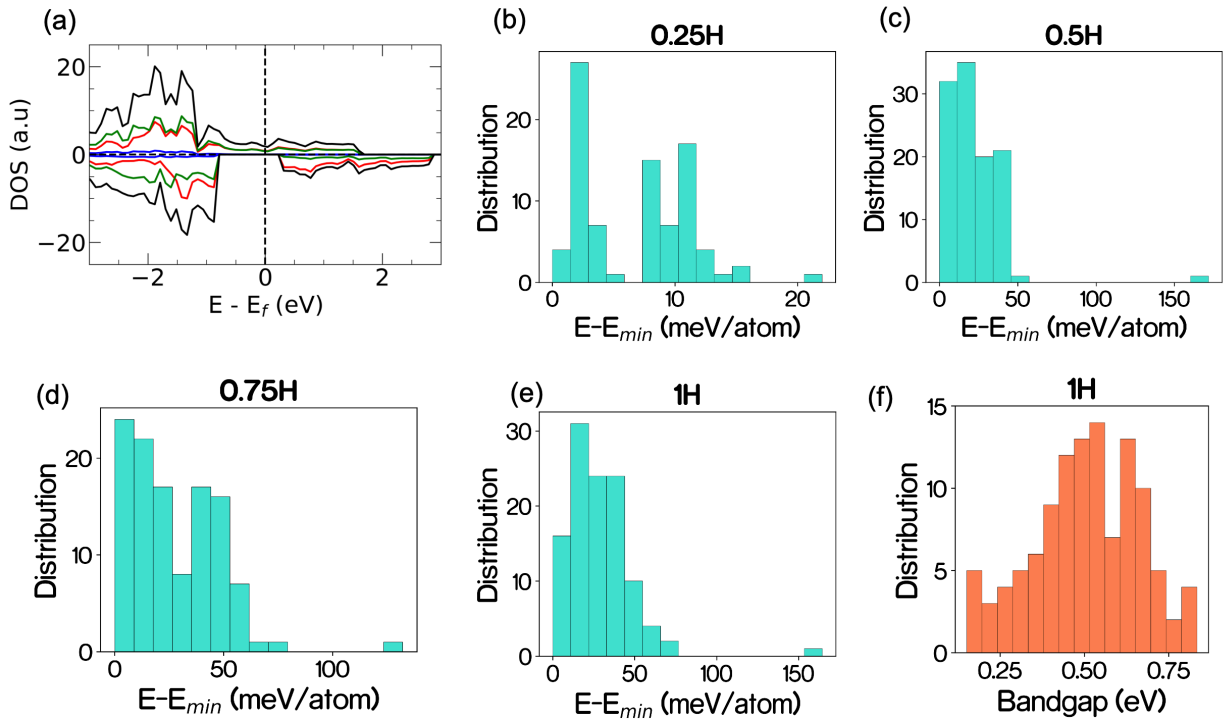

Fig. S3. *(a) The density of states (DOS) of pristine NNO. (b-e) Energy distribution of the MCTS sampled metastable configurations with a doping concentration of 0.25H, 0.5H, 0.75H, and 1H respectively. (f) Band gap distribution of the sampled configurations for a doping concentration of 1H.*

S. 2. First-principles calculations of the electronic density of states and band gap:

The First principal calculations are computed with a DFT+U method using a VASP[3] package. A PBEsol[4] exchange-correlation functional with U_{eff} = 2 eV is used to calculate the structural and electronic properties of pristine NNO and H-doped NNO. This also conforms to the calculation settings previously used to compute the pure metallic state of pure NNO and the H-induced insulating phase of H-NNO. The pseudopotentials Nd_3 (06Sep2000), Ni_pv (Ni_pv 06Sep2000), O (O 08Apr2002), and H (H 15Jun2001) are used for Nd, Ni, O, and H respectively. For the geometry optimization calculations, the plane wave cut-off energy to 500 eV, and the Brillouin zone was sampled at the Γ-point with 6 × 6 × 4 k-point mesh. For calculations of the electronic density of states and band gap, we have used a relatively dense 12 × 12 × 8 k-point mesh sampled at Γ-point. The pristine orthorhombic perovskite (space group Pbnm) NNO structures is used from the Materials Project Database[5].

Reference:

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5. Jain A, Ong SP, Hautier G, Chen W, Richards WD, Dacek S*, et al.* Commentary: The Materials Project: A materials genome approach to accelerating materials innovation. *APL materials* 2013, **1**(1)**:** 011002.