

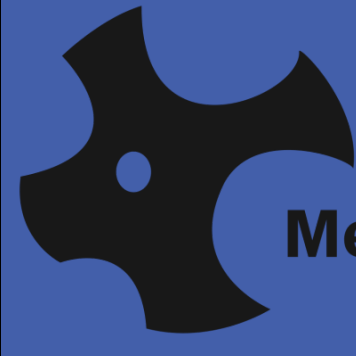
High Throughput Search for New NMC Cathodes

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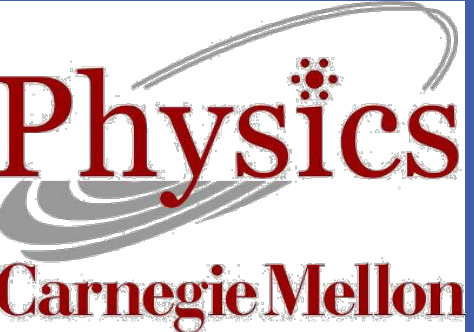
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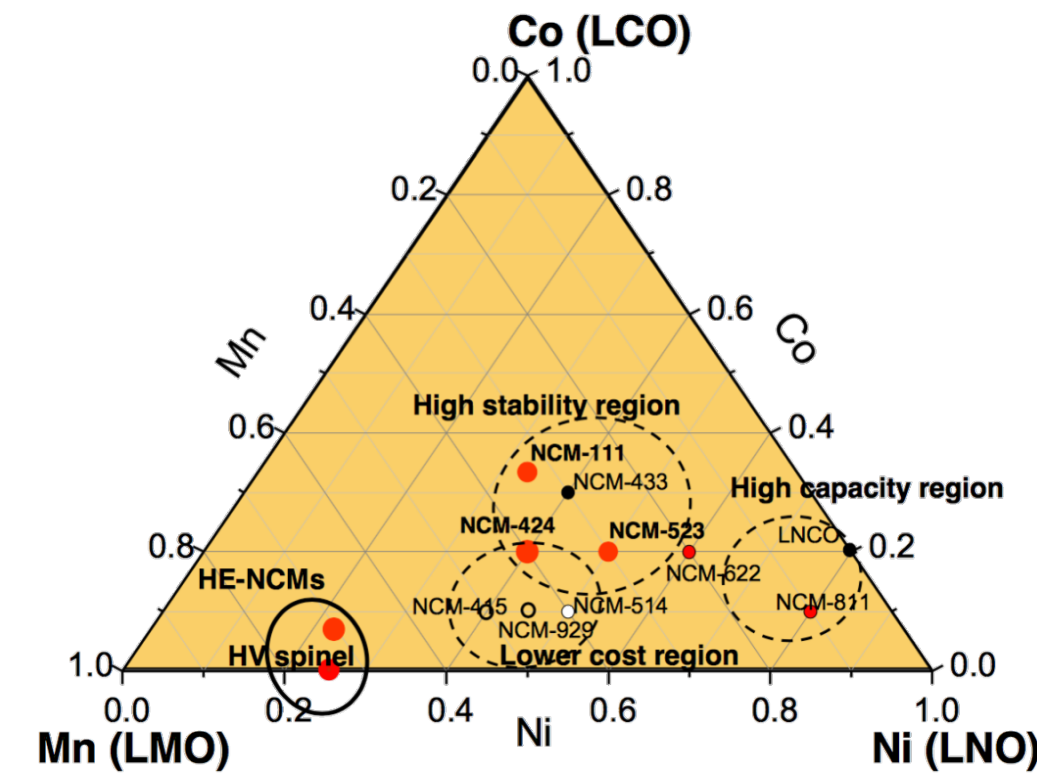


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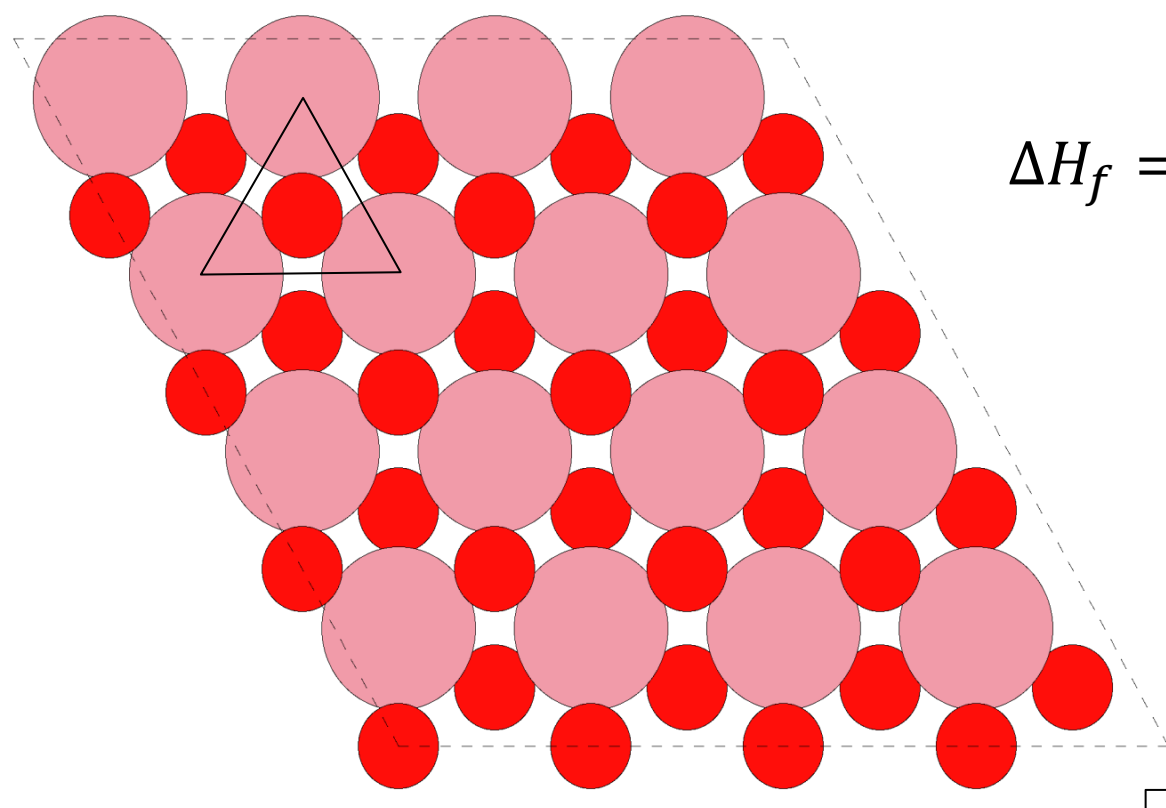
INTRODUCTION

Only a small finite number of $\text{LiNi}_{1-x-y}\text{Mn}_x\text{Co}_y\text{O}_2$ phases have been explored. A high fidelity, exhaustive search with the incorporation of uncertainty estimation is needed.

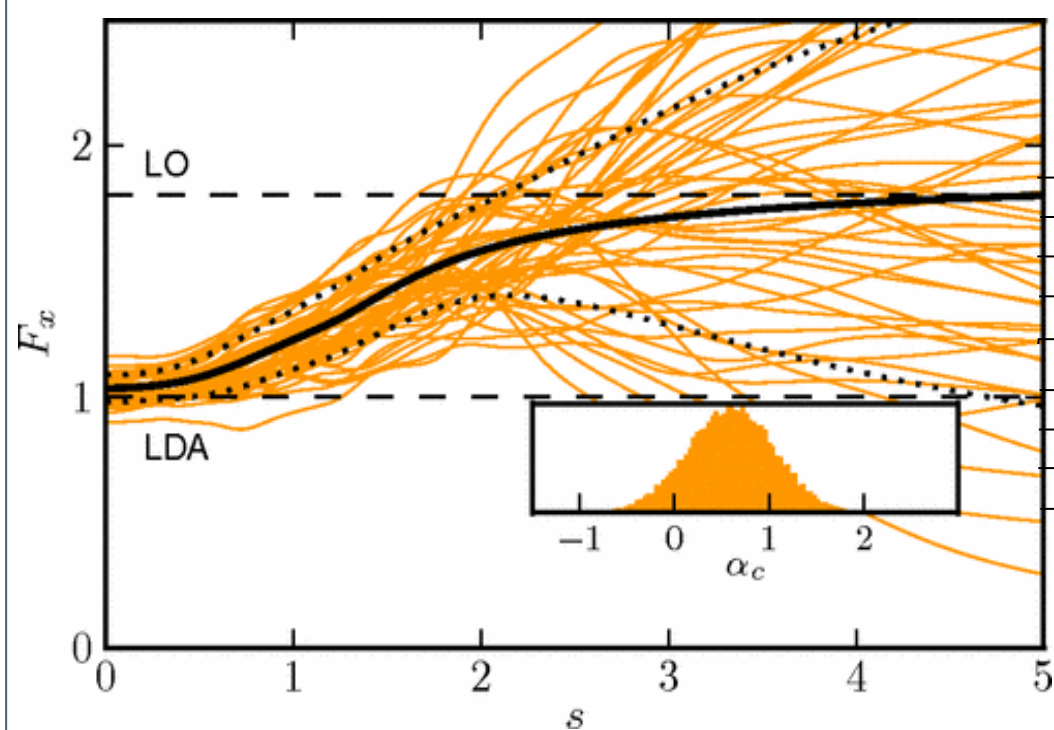


METHODS

We assume a layered structure and model the cation interaction on a triangular lattice. Density functional theory simulations train a reduced order model.



$$\Delta H_f = \sum_{i,x} h_x n_{ix} + \sum_{\langle i,j \rangle, x,y} J_{xy} n_{ix} n_{jy} + \sum_{\langle i,j \rangle, x,y} K_{xy} \vec{S}_{ix} \cdot \vec{S}_{jy}$$

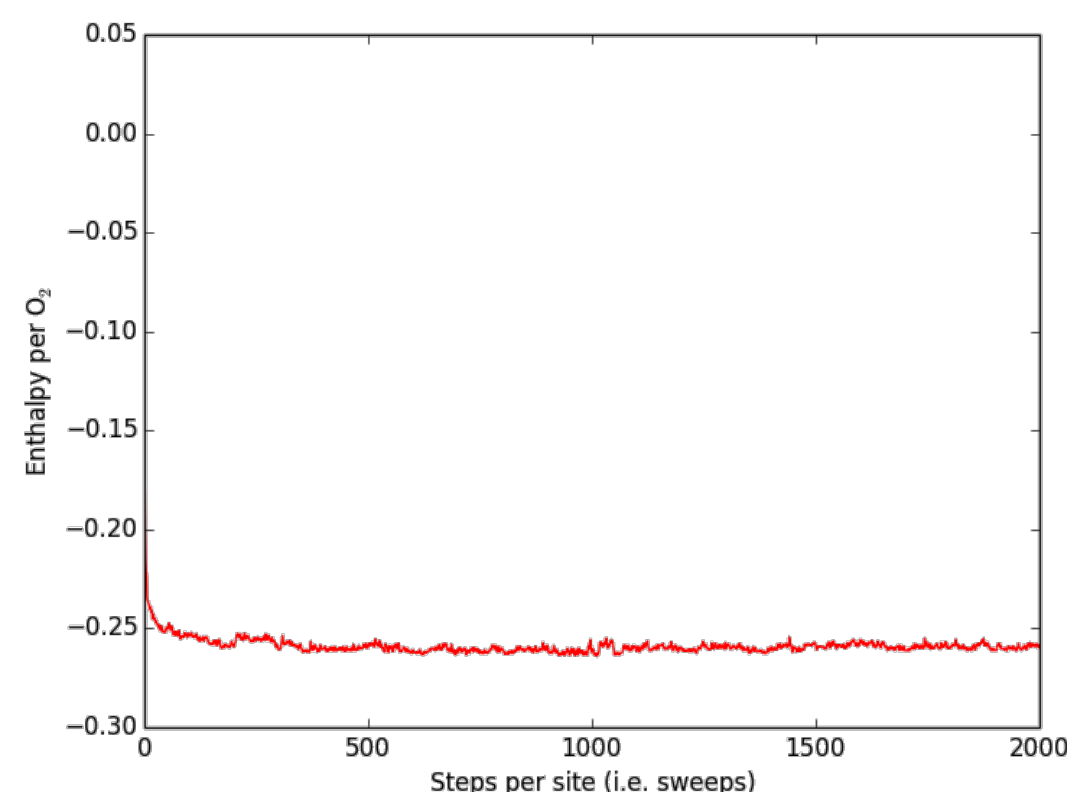


	Max	Min	σ	Value
h_{Ni}	29.8	-11.1	5.8	11.2
h_{Mn}	223.5	-137.9	57.9	46.3
h_{Co}	9.3	-2.7	1.7	2.4
$J_{Ni,Mn}$	-74.9	-222.2	19.3	-151.2
$J_{Mn,Co}$	11.1	-68.5	11.9	-20.1
$J_{Co,Ni}$	47.6	-204.0	34.9	-82.9
$K_{Ni,Ni}$	8.1	-34.6	7.2	-12.0
$K_{Mn,Mn}$	186.1	-122.2	49.4	46.3
$K_{Co,Co}$	105.2	-46.2	22.8	21.7
$K_{Ni,Mn}$	28.6	-27.6	9.2	-3.7
$K_{Mn,Co}$	72.5	-86	21.5	-4.1
$K_{Co,Ni}$	86.2	-166.1	33.9	-42.5

The BEEF-vdW [1] exchange enhancement factor is used to generate an ensemble of energies and therefore an ensemble of fits to the model.

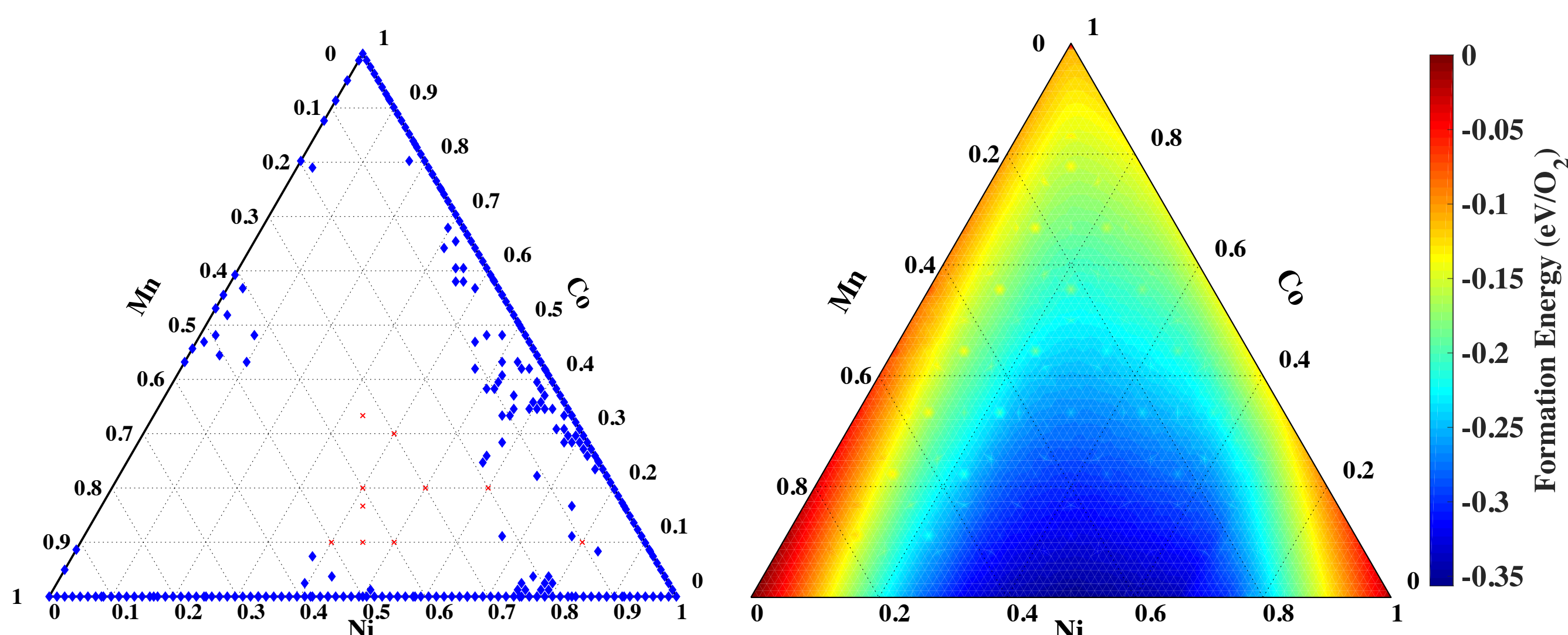
$$\Delta G_f = \Delta H_f - T\Delta S$$

$$\Delta S = -k_B \sum_i p_i \ln p_i$$



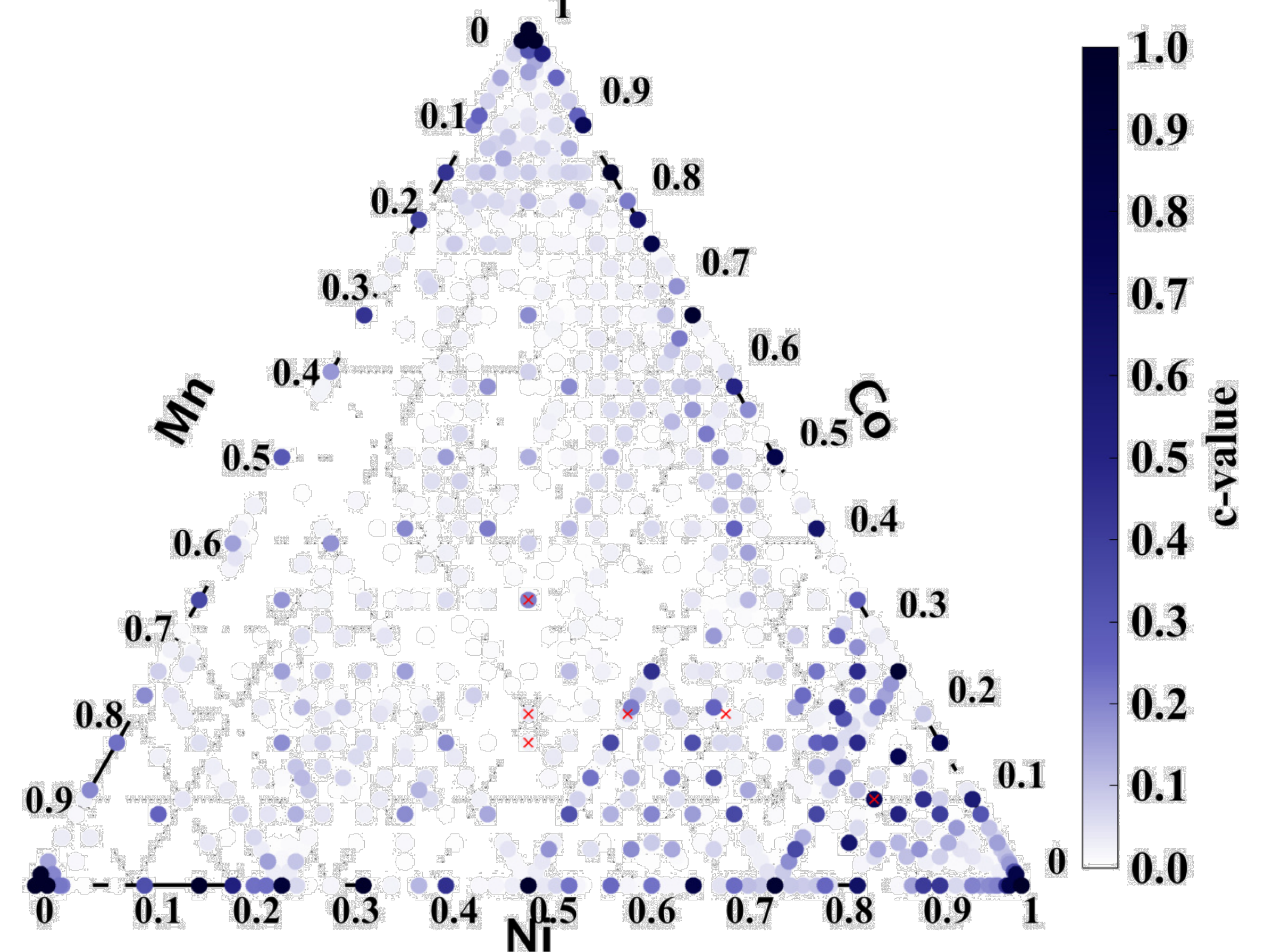
Metropolis Monte Carlo simulations applied to the model generate enthalpy. The statistics of the simulation generate entropy.

RESULTS



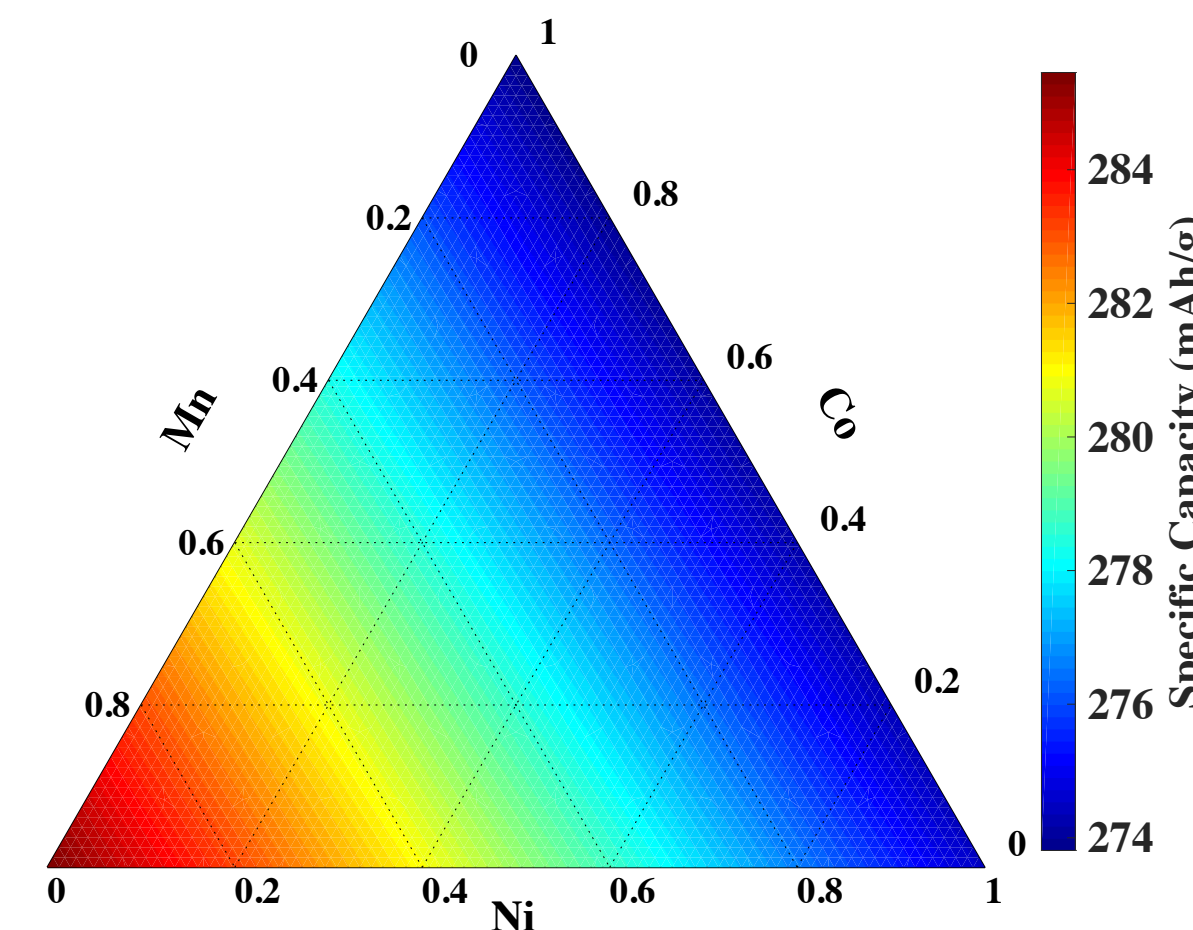
Blue dots represent phases on the convex hull from the main BEEF-vdW functional. The red X's show the currently explored phases.

The ensemble of models generates an ensemble of convex hulls.



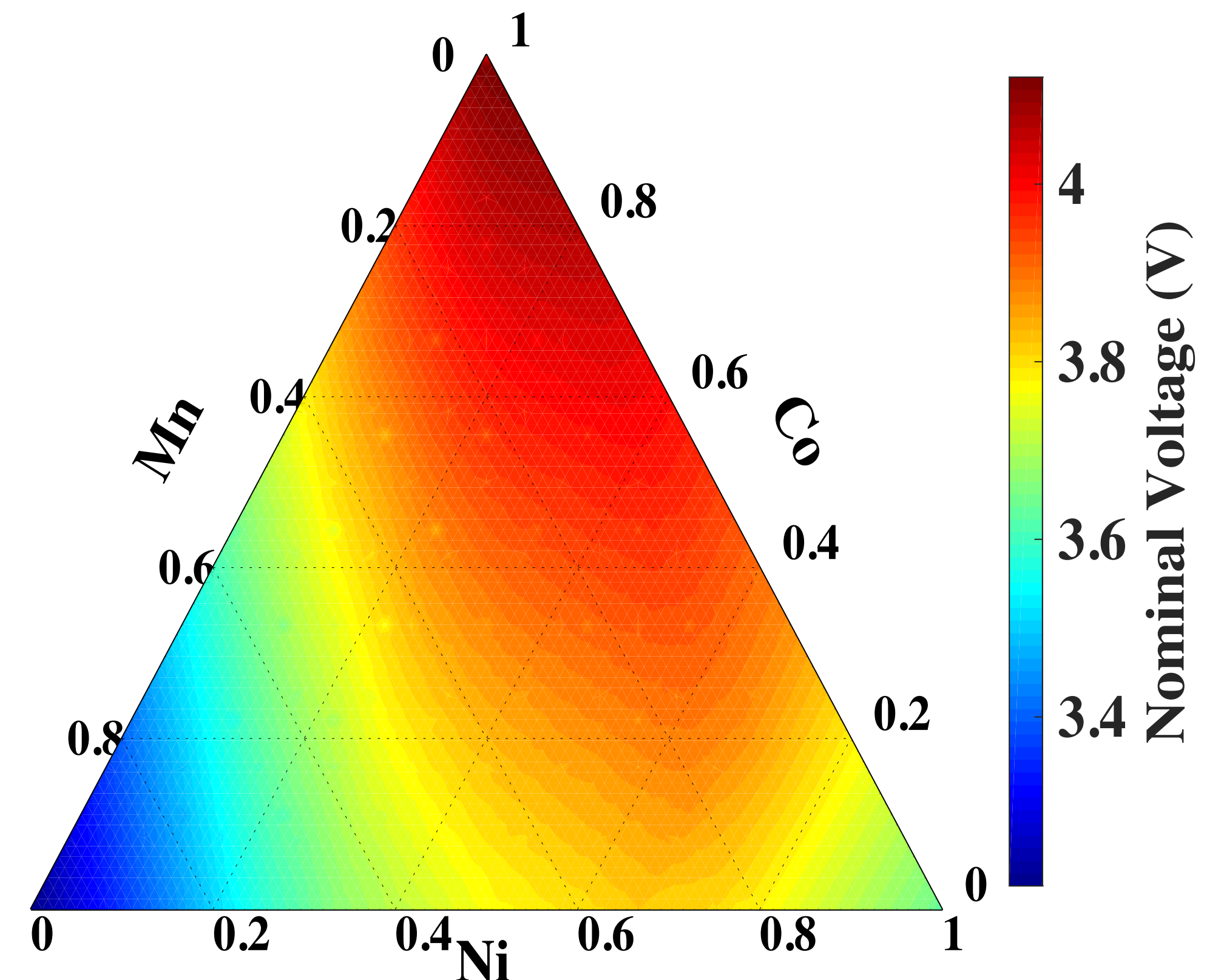
The confidence value (c-value) that a given phase is on the hull is computed as the relative frequency a composition appears on the hull.

INSIGHTS



Experimental Phase	Most likely phase	C-value
111	111	0.449
532	?	
811	10 11	0.425
622	522	0.214
442	552	0.045
541	651	0.141

$$V_{NMC} \approx \frac{-\Delta G_{LiNMC}}{F} + xV_{LiNiO_2} + yV_{LiMnO_2} + (1-x-y)V_{LiCoO_2}$$



Phase	AL1D(V)	Model (V)
111	3.75	3.88
532	3.75	3.76
811	3.76	3.79
622	3.73	3.89

Voltage predictions compare well with average voltage generated from AutoLion-1D simulations

REFERENCES

1. J. Wellendorff, K. T. Lundgaard, A. Møgelhøj, V. Petzold, D. D. Landis, J. K. Nørskov, T. Bligaard, and K. W. Jacobsen, Phys. Rev. B 85, 235149 (2012).
2. G. Houchins, V. Viswanathan Phys. Rev. B 96 134426 (2017).