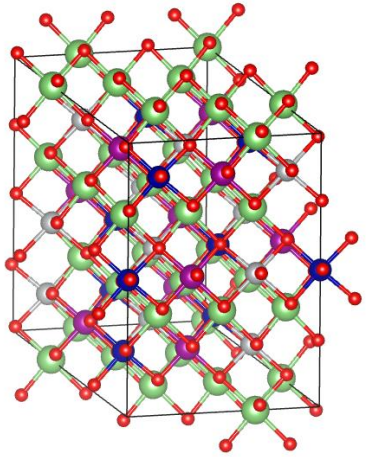
NIST ATOMISTC SIMULATIONS FOR INDUSTRIAL NEEDS, 2020
VIRTUALLY AT GAITHERSBURG, MD, AUGUST 5, 2020

Argonne NATIONAL LABORATORY



MACHINE LEARNING FORCE FIELDS FOR LI-ION CATHODES

JOSHUA J GABRIEL, JUAN GARCIA, NOAH H. PAULSON, JOHN J. LOW, MARIUS STAN, AND HAKIM IDDIR

Argonne National Laboratory
Lemont, IL

CHiMaD

NIST

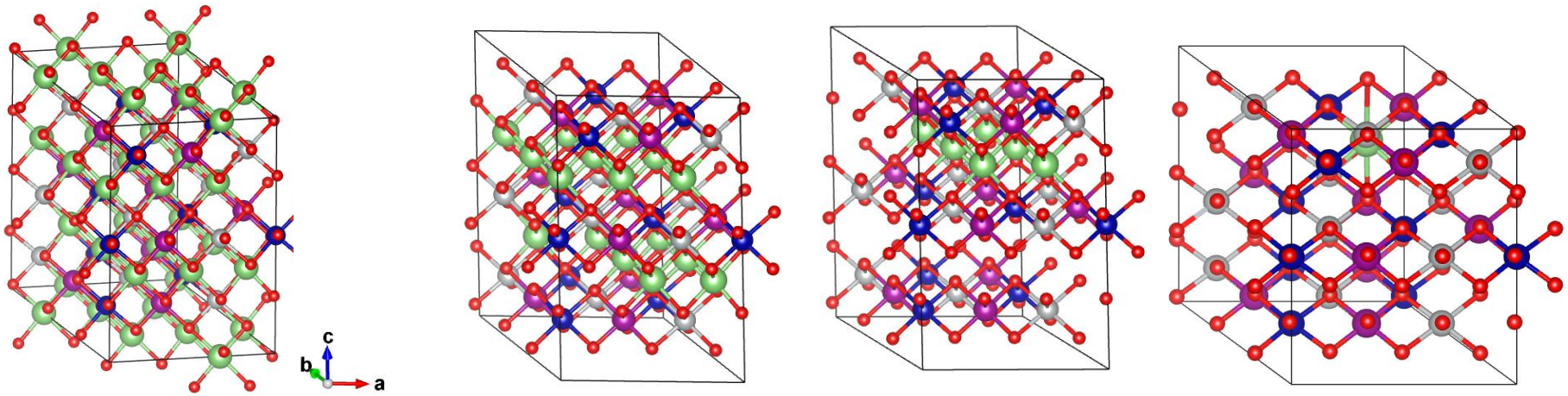
ABSTRACT

NMC (Lithium Nickel Manganese Cobalt Oxide) is a common choice cathode in Lithium ion batteries. However, it is susceptible to fracture at low Li content upon discharging, evidenced by a large change in the *c* lattice constant. We illustrate this collapse through a machine learning force field trained to 1 ps of *ab initio* molecular dynamics (AIMD) for the specific composition NMC-111 that contains equiatomic proportions of nickel, manganese and cobalt. We find the collapse occurs at a Li-content of 0.25% at 298 K in the O3 phase.

BACKGROUND
DELITHIATION OF NMC-111

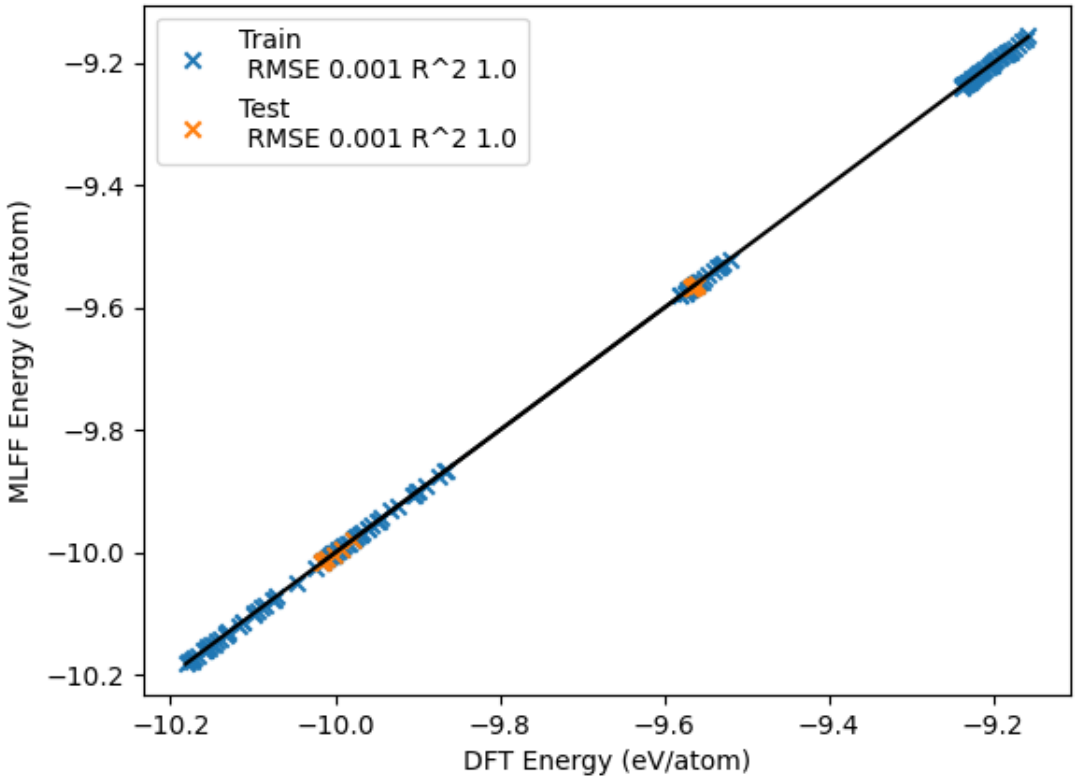
- NMC-111 is a popular cathode that undergoes a well known ‘collapse’ of the interlayer spacing in the *O3* and *O1* structure on delithiation
- We develop a force field to simulate this collapse at operating temperatures for application to other cathodes such as NMC-811

Li=1.0 Ni Co Mn O **Li=0.6 Ni Co Mn O** **Li=0.25 Ni Co Mn O** **Li=0.04 Ni Co Mn O**

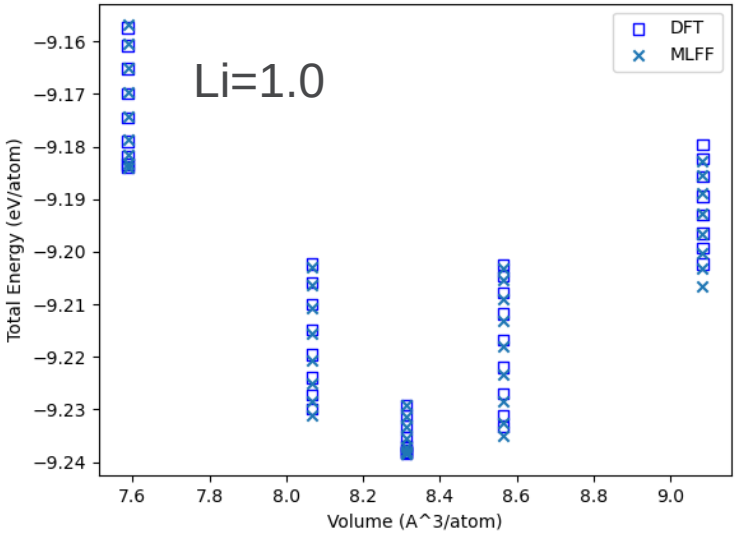


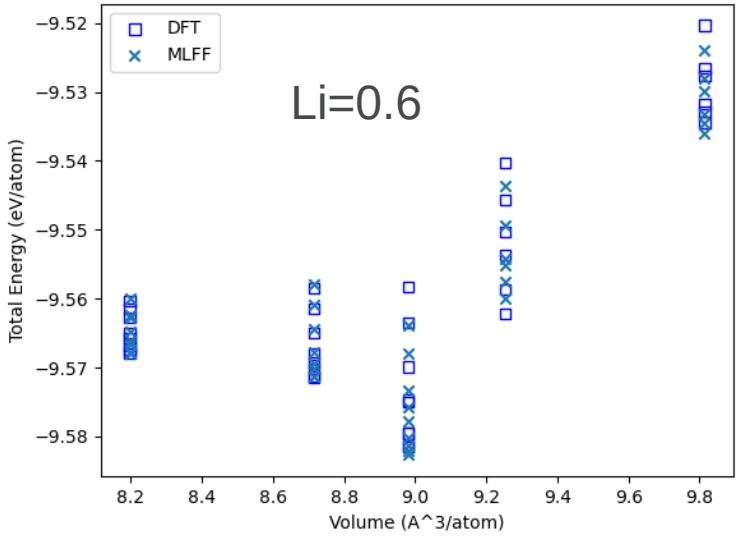
MLFF TRAINED TO 1 PS OF AIMD PREDICTS ENERGIES TO WITHIN 1 MEV/ATOM

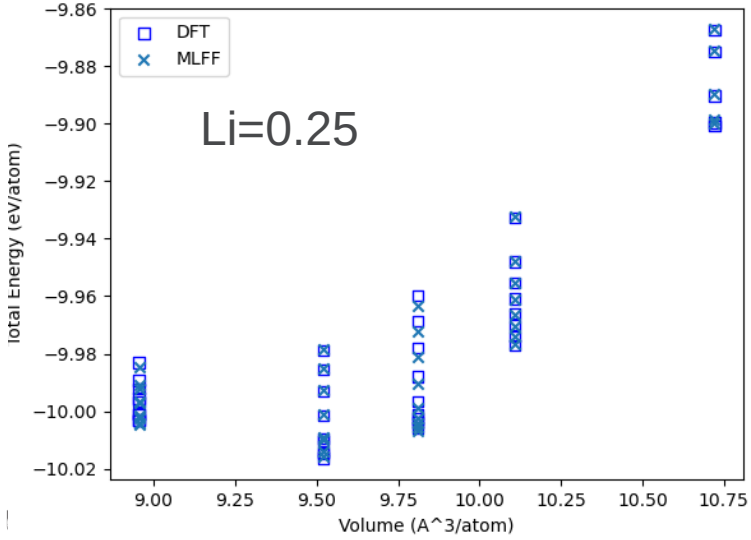
Training over 1 ps AIMD snapshots calculated with DFT level that takes care of van-der Waals interactions and strong correlation in oxides (SCAN+rvv10)

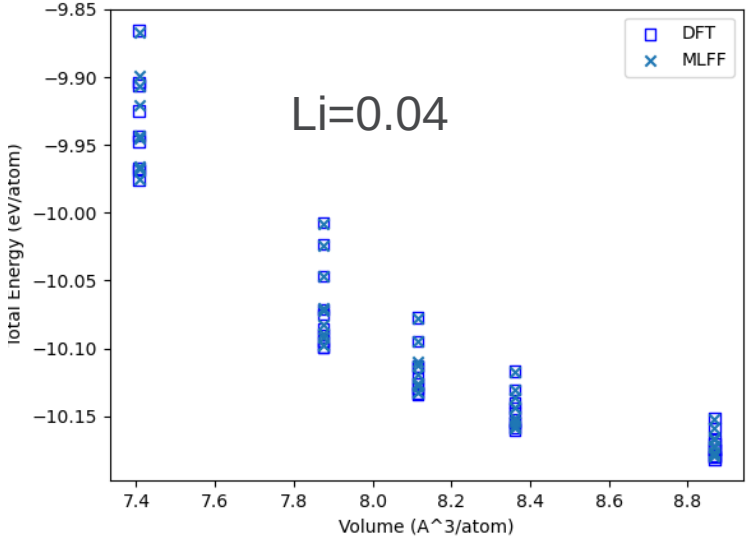


MLFF TRAINED TO 1 PS OF AIMD PREDICTS ENERGIES TO WITHIN 1 MEV/ATOM

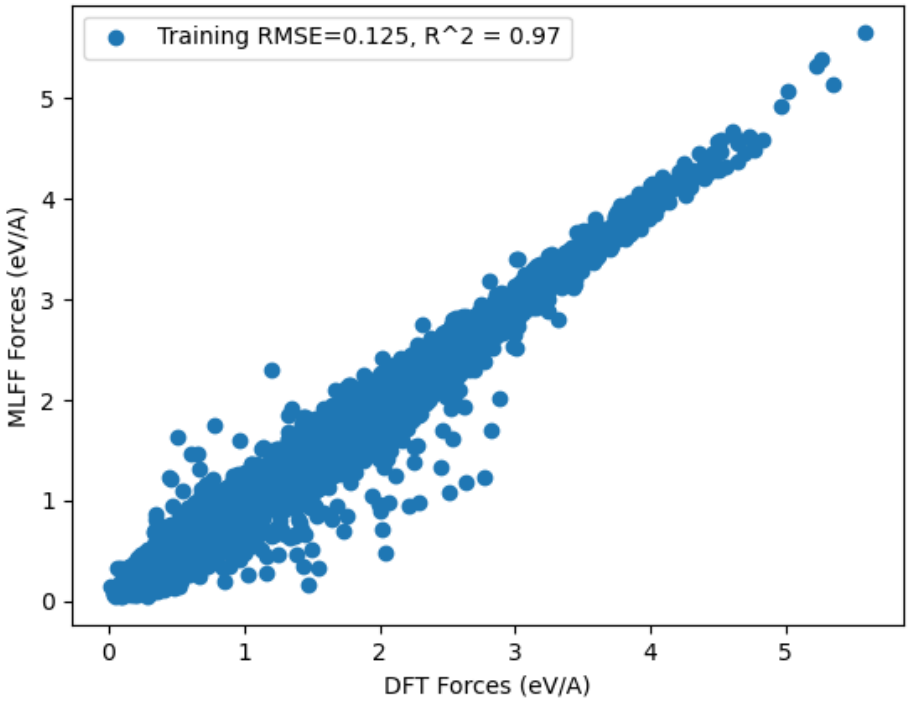








FORCE ERRORS AND PREDICTED LATTICE CONSTANTS AT 298 K



Li-content	C-lattice parameter (MD)	C-lattice parameter (Expt·[1])
1.0	14.51	14.22
0.6	13.94	14.46
0.25	13.79	Experimental transition, mixture of *O1* and *O3* phase
0.04	10.89 (O3 phase)	4.48 (O1 phase)

DeePMD-kit: A deep learning package for many-body potential energy representation and molecular dynamics, *Comput. Phys. Commun.,* 228, 2018, 178-184, https://doi.org/10.1016/j.cpc.2018.03.016

[1]*Chem. Mater.* 2006, 18, 1901-1910

ACKNOWLEDGEMENTS

This research was supported by:

CHiMaD

NIST
National Institute of Standards and Technology
U.S. Department of Commerce

Office of
ENERGY EFFICIENCY &
RENEWABLE ENERGY

Vehicle Technologies Office