A MLIPs and versions in *MaterialsFramework*

Table S1: Comprehensive list of MLIPs currently implemented in *MaterialsFramework*, along with the versions specified for each model. The table includes version identifiers as used in the framework and references to the original publications or official documentation describing each model.

MLIP Name	**Version**	**Reference**
Alignn	v12.2.2024_dft_3d_307k	[36, 37]
AlphaNet†	AlphaNet-oma-v1 AlphaNet-MPtrj-v1	[38]
CHGNet	0.3.0 0.2.0	[39]
DeePMD†	DPA3-v2-OpenLAM DPA3-v2-MPtrj	[40, 41, 42]
EqV2†	eqV2-L-OAM eqV2-M-OAM eqV2-S-OAM eqV2-L-OMat eqV2-M-OMat eqV2-S-OMat eqV2-L-DeNS eqV2-M-DeNS eqV2-S-DeNS eqV2-S	[43]
eSEN†	eSEN-30M-OAM eSEN-30M-OMat eSEN-30M-MP	[44]
GPTFF†	gptff_v2 gptff_v1	[45]
Grace	GRACE-2L-OMAT GRACE-2L-OAM GRACE-1L-OMAT GRACE-1L-OAM GRACE-2L-MP-r6	[46]

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Table S1 – *continued from previous page*

MLIP Name	Version	Reference
	GRACE-2L-MP-r5 GRACE-1L-MP-r6	
HIENet†	HIENet-0	[47]
M3GNet	MP-2021.2.8-PES MatPES-PBE-v2025.1-PES MatPES-r2SCAN-v2025.1-PES 2021.2.8-DIRECT-PES	[48]

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Table S1 – *continued from previous page*

MLIP Name	Version	Reference
MACE	mace_matpes_0 mace_omat_0 mace_mpa_0 mace_mp_0b3 mace_mp_0b2 mace_mp_0b mace_mp_0a	[49]
MatterSim	mattersim-v1.0.0-5m mattersim-v1.0.0-1m	[50]
NewtonNet	ani1 ani1x t1x	[51]
ORB	orb-v3-conservative-20-omat orb-v3-conservative-inf-omat orb-v3-direct-20-omat orb-v3-direct-inf-omat orb-v3-conservative-20-mpa orb-v3-conservative-inf-mpa orb-v3-direct-20-mpa orb-v3-direct-inf-mpa orb-v2 orb-d3-v2 orb-d3-sm-v2 orb-d3-xs-v2 orb-mptraj-only-v2	[52, 53]
PetMad†	v1.1.0 v1.0.1 v1.0.0	[54]
PosEGNN†	pos-egnn.v1-6M	[55]
SevenNet	SevenNet-MF-ompa SevenNet-omat SevenNet-l3i5	[56, 57]

Continued on next page

Table S1 – *continued from previous page*

MLIP Name	Version	Reference
SevenNet-0		

† These models require manual download of their respective checkpoint or parameter files due to licensing or distribution restrictions. Please refer to the official repositories for instructions on obtaining and configuring these files.

B Example of D0$_{19}$ structure for SQS construction

The input file of the D0$_{19}$ structure `rndstr.skel` is:

```
5.305 5.305 4.242 90 90 120   'Coordinate system
1 0 0
0 1 0
0 0 1                          'Primitive unit cell vectors
0.3333 0.6667 0.25 b           'Atom sites in the cell for sublattice 'b'
0.6667 0.3333 0.75 b
0.8333 0.6667 0.25 a           'Atom sites in the cell for sublattice 'a'
0.8333 0.1667 0.25 a
0.1667 0.8333 0.25 a
0.1667 0.3333 0.75 a
0.6667 0.8333 0.75 a
0.1667 0.8333 0.75 a
```

where a and b indicate the different sublattices. The `sqsgen.in` is:

```
level=0 a=1          b=1
level=1 a=0.5,0.5    b=1
level=1 a=1          b=0.5,0.5
level=2 a=0.5,0.5    b=0.5,0.5
level=3 a=0.75,0.25 b=1
level=3 a=1          b=0.75,0.25
```

In this case, for level=2, we have both a and b sublattice occupied by two elements, with $x^a(A) = x^a(B) = 0.5$, $x^b(C) = x^b(D) = 0.5$, where A, B, C and D are the elements in sublattice a and b. The output file `bestsqs.out` is:

```
5.305000 0.000000 0.000000
-2.652500 4.594265 0.000000
0.000000 0.000000 4.242000
0.000000 -1.000000 0.000000
0.000000 0.000000 -2.000000
2.000000 0.000000 0.000000
0.333300 -0.333300 -0.750000 b_A
1.333300 -0.333300 -0.750000 b_B
0.333300 -0.333300 -1.750000 b_B
1.333300 -0.333300 -1.750000 b_A
0.666700 -0.666700 -0.250000 b_B
1.666700 -0.666700 -0.250000 b_A
0.666700 -0.666700 -1.250000 b_B
1.666700 -0.666700 -1.250000 b_A
0.833300 -0.333300 -0.750000 a_B
1.833300 -0.333300 -0.750000 a_A
0.833300 -0.333300 -1.750000 a_B
1.833300 -0.333300 -1.750000 a_A
0.833300 -0.833300 -0.750000 a_A
1.833300 -0.833300 -0.750000 a_B
0.833300 -0.833300 -1.750000 a_A
1.833300 -0.833300 -1.750000 a_B
```

```
0.333300 -0.833300 -0.750000 a_A
1.333300 -0.833300 -0.750000 a_B
0.333300 -0.833300 -1.750000 a_B
1.333300 -0.833300 -1.750000 a_A
0.166700 -0.666700 -0.250000 a_B
1.166700 -0.666700 -0.250000 a_B
0.166700 -0.666700 -1.250000 a_A
1.166700 -0.666700 -1.250000 a_A
0.666700 -0.166700 -0.250000 a_A
1.666700 -0.166700 -0.250000 a_B
0.666700 -0.166700 -1.250000 a_A
1.666700 -0.166700 -1.250000 a_B
0.166700 -0.166700 -0.250000 a_B
1.166700 -0.166700 -0.250000 a_B
0.166700 -0.166700 -1.250000 a_A
1.166700 -0.166700 -1.250000 a_A
```

for the SQS of $x_A = x_B = 0.5$ in both sublattices.

C Ab-initio data of energy and vibrational entropy of pure metallic elements

Table S2: Ab-initio data of energy and vibrational entropy of pure metallic elements

Element	FCC_A1			BCC_A2			HCP_A3		
	Unstable	Energy (eV)	Vibrational Entropy	Unstable	Energy (eV)	Vibrational Entropy	Unstable	Energy (eV)	Vibrational Entro
Ag		-2.72433	-86.1889		-2.61169	-84.5878		-5.4422	-172.172
Al		-3.74135	-88.2663	✓	-3.69447	-87.8629		-7.42034	-176.257
Au		-3.22782	-85.3757		-3.207595	-86.3942		-6.44402	-170.539
Co		-7.02531	-88.0018	✓	-6.9442	-87.7826		-14.0868	-176.362
Cr	✓	-9.32554	-88.8351		-9.49173	-88.8181	✓	-18.5863	-176.763
Cu		-3.73145	-87.5273		-3.69644	-87.2028		-7.44798	-174.943
Fe		-8.08568	-88.3182		-8.24337	-88.4823		-16.1316	-176.131
Hf		-9.88344	-85.6048		-9.7776	-85.7277		-19.9156	-172.170
In		-2.55594	-84.4606		-2.54688	-84.2564		-5.10406	-168.515
Ir		-8.85517	-87.0129	✓	-8.23357	-87.2042		-17.5588	-173.746
Mg		-1.49287	-87.5432		-1.47669	-87.3609		-3.01029	-175.256
Mn		-8.90193	-87.8455		-8.82125	-87.5310		-17.8653	-175.886
Mo	✓	-10.711	-87.0004		-10.9427	-87.9353	✓	-21.4129	-174.970
Nb	✓	-10.0079	-87.0338		-10.2089	-87.1036	✓	-10.1419	-173.937
Ni		-5.47533	-88.1199	✓	-5.38133	-87.9685		-10.9017	-175.971
Os		-11.0983	-87.096	✓	-11.0050	-87.8551		-22.478	-174.983
Pb		-3.5709	-83.8365	✓	-3.5358	-84.1784		-7.11132	-167.472
Pd		-5.21951	-87.0289		-5.17659	-86.7851		-10.3838	-173.718
Pt		-6.10114	-86.4436	✓	-6.05414	-86.6885		-12.0903	-172.131
Re		-12.2638	-85.9419	✓	-12.2426	-87.3593		-24.7992	-173.732
Rh		-7.28059	-87.5303	✓	-7.06104	-87.2504		-14.4864	-175.004
Ru		-9.14488	-87.5171	✓	-9.0875	-87.5499		-18.5230	-176.085
Sc		-6.15546	-87.1803	✓	-6.1522	-89.4009		-12.4064	-178.000
Sn		-3.77427	-84.2970		-3.76344	-84.2074		-7.55217	-168.786
Ta	✓	-11.6899	-86.5449		-11.8579	-86.3414	✓	-23.3963	-172.704
Ti		-7.7081	-87.2747	†				-15.5295	-175.495
V	✓	-8.90428	-87.8334		-8.94585	-87.6379	✓	-17.8918	-175.53
W	✓	-12.7685	-87.3763		-13.008	-87.4066	✓	-25.7543	-174.269
Y		-6.39954	-86.0638		-6.30011	-85.6655		-12.8496	-172.246
Zn		-1.08594	-86.1428		-1.02165	-86.0296	✓	-2.19499	-172.505
Zr		-8.46931	-86.3133		-8.42381	-86.2414		-17.0165	-173.524

† Dynamically Stabilized (DOI: 10.1103/PhysRevB.95.064101);
* The ✓ in "Unstable" indicates the phase is mechanically unstable and the "inflection detection" method is applied;
** The "Energy (eV)" is the energy per primitive cell (1 atom per cell for FCC_A1 and BCC_A2, 2 atoms per cell for HCP_A3) at 0 K;
*** The "Vibrational Entropy" is given per primitive cell, in units of "Boltzmann constant per cell" – consistent with the convention in ATAT and can be directly used to replace `svib_ht`.

D LAMMPS input script for liquid-phase MD calculations using MLIPs

```
variable        s equal step
variable        t equal temp
variable        v equal vol
variable        e equal pe
variable        p equal press

units           metal
dimension       3
boundary        p p p
atom_style      atomic

read_data       <STRUCTURE>

pair_style      gnnp <ML-GNNP PATH>
pair_coeff      * *  <MLIP> <VERSION> <ELEMENTS>

timestep        0.001  # 1 fs

# Minimization
reset_timestep  0
thermo          10
thermo_style    custom step temp etotal pe lx ly lz press pxx pyy pzz

min_style       cg
minimize        1e-25 1e-25 5000 10000

reset_timestep  0
velocity        all create <TEMPERATURE> <TEMPERATURE SEED> mom yes rot no

# Equilibration: NPT
fix             1 all npt temp <TEMPERATURE> <TEMPERATURE> $(10.0*dt) iso 0.0 0.0 $
    (100.0*dt)
thermo          100
thermo_style    custom step temp press pe ke etotal lx ly lz
run             30000  # 30 ps
unfix           1

# Production: NVT
fix             2 all nvt temp <TEMPERATURE> <TEMPERATURE> $(100.0*dt)
fix             Eavg all ave/time 10 3000 30000 etotal file energy ave one
fix_modify      Eavg title1 "" title2 "" title3 ""
thermo          100
thermo_style    custom step temp press pe ke etotal lx ly lz
run             30000  # 30 ps
unfix           2
unfix           Eavg
```