Supporting information

Stability and Transferability of Machine Learning Force Field for Molecular Dynamics Applications

*Salatan Duangdangchote, Dwight S. Seferos, and Oleksandr Voznyy**

Prof. O. Voznyy, S. Duangdangchote

Department of Physical and Environmental Sciences, University of Toronto (Scarborough), 1065 Military Trail, M1C 1A4 Toronto, Ontario, Canada

Prof. D. S. Seferos

Department of Chemistry, University of Toronto, Toronto, ON M5S 3H6, Canada

Table S1 Comparison of MLFF-MD computation results with AIMD reference data of $Li_{10}GeP_2S_{12}$ for energy, forces, and radial distribution functions (RDF) across various temperatures

Model	***MAE***	**Temperature**					*Average*
		800 K	900 K	1000 K	1100 K	1200K	
CGCNN	Energy	0.105	0.106	0.116	0.136	0.166	0.126
	Forces	0.051	0.056	0.063	0.167	0.072	0.082
	RDF	0.007	0.008	0.087	0.282	0.601	0.197
SchNet	Energy	0.079	0.055	0.616	0.084	0.120	0.191
	Forces	0.024	0.028	0.031	0.033	0.037	0.031
	RDF	0.007	0.004	0.003	0.003	0.009	0.005
DimeNet	Energy	0.027	0.033	0.038	0.047	0.053	0.030
	Forces	0.011	0.013	0.014	0.015	0.017	0.014
	RDF	0.003	0.004	0.003	0.002	0.003	0.003
DimeNet++	Energy	0.022	0.025	0.029	0.034	0.038	0.030
	Forces	0.008	0.009	0.010	0.011	0.012	0.010
	RDF	0.003	0.006	0.004	0.002	0.003	0.004
GNS-TAT	Energy	0.074	0.046	0.041	0.056	0.083	0.060
	Forces	0.011	0.013	0.014	0.015	0.017	0.014
	RDF	0.037	0.003	0.005	0.154	0.078	0.055
DeeperGATGNN	Energy	0.095	0.091	0.103	0.133	0.166	0.118
	Forces	0.042	0.047	0.051	0.055	0.059	0.051
	RDF	140.703	34.883	112.352	35.528	101.464	84.986
SCN	Energy	0.028	0.033	0.034	0.038	0.042	0.035
	Forces	0.006	0.007	0.007	0.008	0.009	0.007
	RDF	0.003	0.006	0.003	0.003	0.003	0.004
eSCN	Energy	0.061	0.052	0.046	0.044	0.041	0.049
	Forces	0.007	0.008	0.009	0.009	0.010	0.009
	RDF	0.004	0.006	0.004	0.004	0.003	0.004
ForceNet	Energy	N/A	N/A	N/A	N/A	N/A	–
	Forces	0.020	0.022	0.024	0.026	0.287	0.076
	RDF	0.274	0.272	0.302	0.279	0.282	0.282
Equiformer	Energy	0.105	0.096	0.094	0.104	0.019	0.084
	Forces	0.012	0.013	0.014	0.015	0.016	0.014
	RDF	0.005	0.006	0.005	0.003	0.003	0.004
LeftNet	Energy	0.039	0.045	0.047	0.052	0.058	0.048
	Forces	0.009	0.011	0.013	0.014	0.015	0.012
	RDF	0.003	0.005	0.004	0.003	0.003	0.004

Table S2 Comparison of MLFF-MD computation results with AIMD reference data of Li_3PS_4 for energy, forces, and radial distribution functions (RDF) across various temperatures

Model	***MAE***	**Temperature**					*Average*
		800 K	900 K	1000 K	1100 K	1200K	
CGCNN	Energy	0.105	0.106	0.116	0.136	0.166	0.126
	Forces	0.051	0.056	0.063	0.167	0.072	0.082
	RDF	0.007	0.008	0.087	0.282	0.601	0.197
SchNet	Energy	0.079	0.055	0.616	0.084	0.120	0.191
	Forces	0.024	0.028	0.031	0.033	0.037	0.031
	RDF	0.007	0.004	0.003	0.003	0.009	0.005
DimeNet	Energy	0.027	0.033	0.038	0.047	0.053	0.030
	Forces	0.011	0.013	0.014	0.015	0.017	0.014
	RDF	0.003	0.004	0.003	0.002	0.003	0.003
DimeNet++	Energy	0.022	0.025	0.029	0.034	0.038	0.030
	Forces	0.008	0.009	0.010	0.011	0.012	0.010
	RDF	0.003	0.006	0.004	0.002	0.003	0.004
GNS-TAT	Energy	0.074	0.046	0.041	0.056	0.083	0.060
	Forces	0.011	0.013	0.014	0.015	0.017	0.014
	RDF	0.037	0.003	0.005	0.154	0.078	0.055
DeeperGATGNN	Energy	0.095	0.091	0.103	0.133	0.166	0.118
	Forces	0.042	0.047	0.051	0.055	0.059	0.051
	RDF	140.703	34.883	112.352	35.528	101.464	84.986
SCN	Energy	0.028	0.033	0.034	0.038	0.042	0.035
	Forces	0.006	0.007	0.007	0.008	0.009	0.007
	RDF	0.003	0.006	0.003	0.003	0.003	0.004
eSCN	Energy	0.061	0.052	0.046	0.044	0.041	0.049
	Forces	0.007	0.008	0.009	0.009	0.010	0.009
	RDF	0.004	0.006	0.004	0.004	0.003	0.004
ForceNet	Energy	N/A	N/A	N/A	N/A	N/A	–
	Forces	0.135	0.139	0.146	0.148	0.152	0.144
	RDF	0.370	0.479	0.410	0.431	0.377	0.413
Equiformer	Energy	74.168	74.154	74.153	74.179	74.145	74.160
	Forces	0.096	0.100	0.086	0.099	0.087	0.094
	RDF	1.294	0.300	0.451	0.380	0.022	0.489
LeftNet	Energy	73.324	73.333	73.173	73.367	73.341	73.308
	Forces	0.090	0.093	0.081	0.093	0.083	0.088
	RDF	0.080	0.050	0.054	0.042	0.038	0.053

Table S3 Comparison of MLFF-MD computation results of GNN model with AIMD reference data of Li_4GeS_4 for energy, forces, and radial distribution functions (RDF) across various temperatures

Model	***MAE***	**Temperature**					*Average*
		800 K	900 K	1000 K	1100 K	1200K	
CGCNN	Energy	45.356	45.342	45.706	45.686	45.801	45.578
	Forces	0.515	0.524	0.544	0.554	0.566	0.541
	RDF	1.077	1.274	1.369	1.607	1.637	1.393
SchNet	Energy	56.285	56.362	56.385	56.455	56.548	56.407
	Forces	0.175	0.179	0.194	0.198	0.213	0.192
	RDF	N/A	N/A	N/A	N/A	N/A	–
DimeNet	Energy	61.553	61.552	61.493	61.517	61.426	61.508
	Forces	0.082	0.084	0.086	0.089	0.091	0.086
	RDF	0.018	0.018	0.015	0.012	0.013	0.015
DimeNet++	Energy	61.977	61.976	61.938	61.966	61.896	61.951
	Forces	0.082	0.084	0.086	0.089	0.092	0.087
	RDF	0.018	0.017	0.013	0.009	0.012	0.014
GNS-TAT	Energy	60.916	60.721	60.382	60.209	59.844	60.414
	Forces	0.219	0.228	0.236	0.245	0.252	0.236
	RDF	0.037	0.035	0.050	0.041	0.049	0.042
DeeperGATGNN	Energy	56.034	55.914	55.629	55.506	55.104	55.637
	Forces	0.285	0.304	0.334	0.349	0.383	0.331
	RDF	0.111	0.108	0.087	0.097	0.483	0.177
SCN	Energy	62.114	62.125	62.137	62.186	62.146	62.142
	Forces	0.080	0.082	0.083	0.085	0.087	0.083
	RDF	0.032	0.025	0.031	0.017	0.015	0.024
eSCN	Energy	61.896	61.907	61.909	61.954	61.919	61.917
	Forces	0.080	0.082	0.083	0.085	0.087	0.083
	RDF	0.046	0.041	0.051	0.037	0.029	0.041
ForceNet	Energy	N/A	N/A	N/A	N/A	N/A	–
	Forces	0.094	0.098	0.105	0.109	0.117	0.105
	RDF	0.382	0.398	0.345	0.267	0.342	0.347
Equiformer	Energy	62.142	62.174	62.167	62.230	62.191	62.181
	Forces	0.082	0.084	0.087	0.089	0.091	0.087
	RDF	0.420	0.049	0.291	0.076	0.164	0.200
LeftNet	Energy	61.744	61.746	61.796	61.837	61.871	61.799
	Forces	0.077	0.079	0.081	0.083	0.086	0.081
	RDF	0.017	0.015	0.015	0.008	0.010	0.013

Table S4 Predicting the negative logarithm of the diffusivity via molecular dynamics of $Li_{10}GeP_2S_{12}$ across different temperatures using machine learning force field against AIMD reference data

Model	**Temperatures**					R^2	*MAE*
	800 K	900 K	1000 K	1100 K	1200 K		
AIMD	2.731	4.114	4.753	5.610	8.013	0.942	
CGCNN	1.843	1.555	3.065	4.275	9.153	0.786	1.782
SchNet	2.099	2.170	3.377	4.336	4.958	0.955	1.394
DimeNet	1.616	1.800	2.487	4.053	5.400	0.921	0.851
DimeNet++	1.796	1.788	3.337	4.336	4.958	0.955	1.394
GNS-TAT	1.692	2.960	2.888	3.473	6.128	0.814	1.186
DeeperGATGNN	0.780	1.070	0.875	2.556	0.559	0.043	3.436
SCN	1.944	2.356	2.586	4.487	6.170	0.884	1.253
eSCN	1.463	1.845	2.860	1.776	2.751	0.400	1.793

ForceNet	0.000	0.000	0.000	0.000	0.000	–	–
Equiformer	2.023	2.214	2.748	4.208	5.720	0.903	1.402
LeftNet	1.592	1.439	1.469	1.505	4.270	0.478	1.619

Table S5 Predicting the negative logarithm of the diffusivity via molecular dynamics of Li_3PS_4 across different temperatures using machine learning force field against AIMD reference data

Model	**Temperatures**					R^2	*MAE*
	800 K	900 K	1000 K	1100 K	1200 K		
AIMD	1.561	3.385	6.938	6.795	11.663	0.928	
CGCNN	0.261	0.338	1.161	5.292	8.492	0.860	4.462
SchNet	N/A	N/A	N/A	N/A	N/A	–	–
DimeNet	1.813	4.263	6.264	7.384	11.437	0.966	0.178
DimeNet++	2.268	3.864	5.928	8.810	12.456	0.972	0.374
GNS-TAT	0.032	0.172	0.920	1.092	1.574	0.958	5.607
DeeperGATGNN	1.349	1.253	2.019	1.561	2.359	0.617	4.556
SCN	2.570	3.347	4.715	5.777	9.823	0.889	0.328
eSCN	0.156	0.643	1.397	4.492	4.108	0.856	4.336
ForceNet	0.000	0.000	0.000	0.000	0.000	–	–

Equiformer	2.023	2.214	2.748	4.208	5.720	0.903	1.402
LeftNet	1.592	1.439	1.469	1.505	4.270	0.478	1.619

Table S6 Predicting the negative logarithm of the diffusivity via molecular dynamics of Li_4GeS_4 across different temperatures using machine learning force field against AIMD reference data

Model	**Temperatures**					R^2	*MAE*
	800 K	900 K	1000 K	1100 K	1200 K		
AIMD	0.005	0.229	1.774	2.805	5.305	0.927	
CGCNN	2.654	2.850	4.734	3.644	4.387	0.542	2.783
SchNet	N/A	N/A	N/A	N/A	N/A	–	–
DimeNet	0.045	0.524	0.823	2.721	5.360	0.863	1.911
DimeNet++	0.119	0.317	2.450	2.688	5.341	0.910	1.911
GNS-TAT	0.000	0.000	0.000	0.003	0.234	0.510	2.398
DeeperGATGNN	0.082	0.174	0.254	0.554	0.488	0.862	2.302
SCN	0.000	0.000	0.395	1.235	3.099	0.811	2.150
eSCN	0.000	0.000	0.000	0.556	1.991	0.692	2.257
ForceNet	0.000	0.000	0.000	0.000	0.000	–	–

Equiformer	0.381	0.624	2.895	3.352	4.919	0.947	1.788
LeftNet	0.376	0.285	1.821	2.850	4.350	0.934	1.788

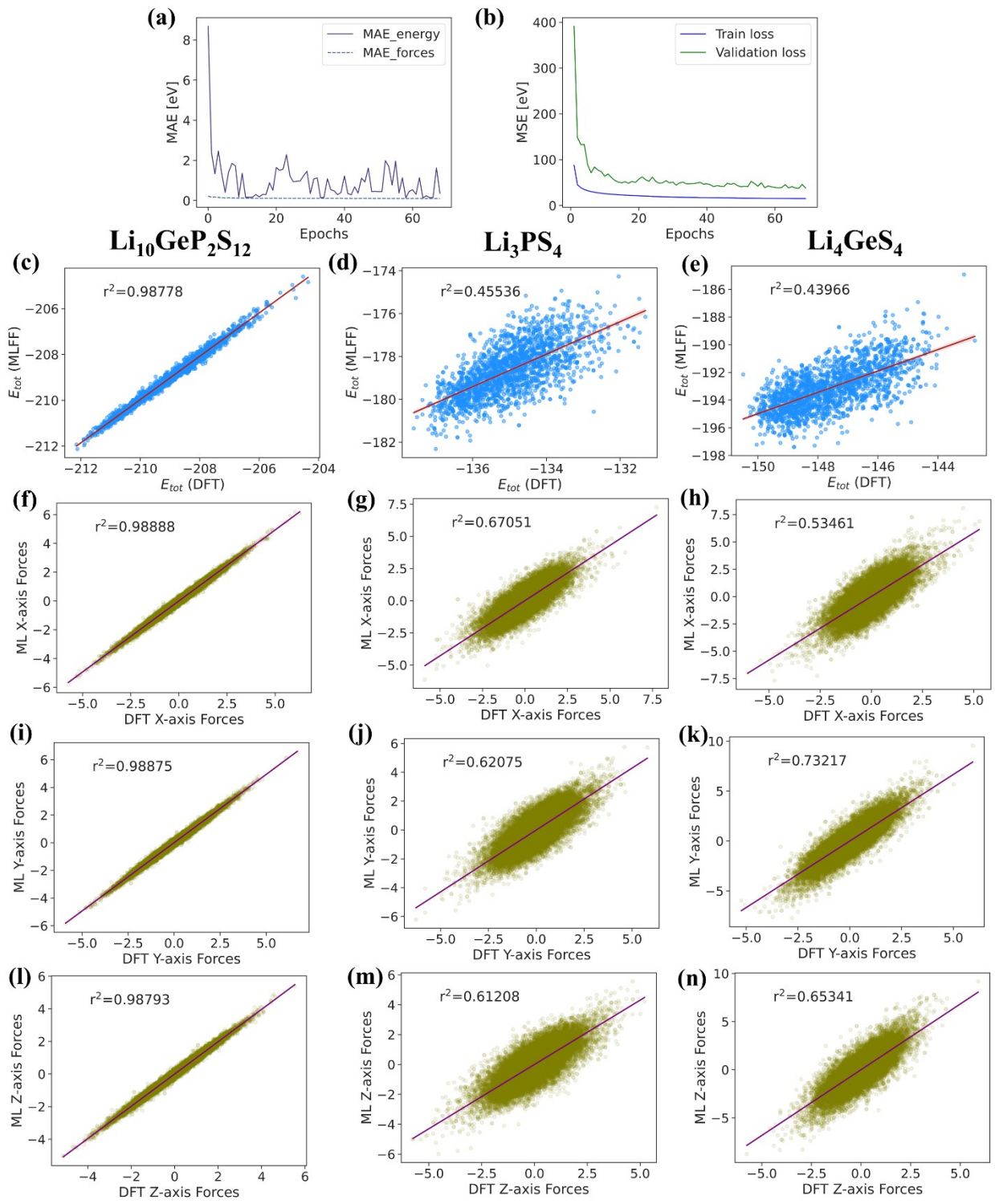

Figure S1 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of CGCNN model against AIMD reference data for different materials

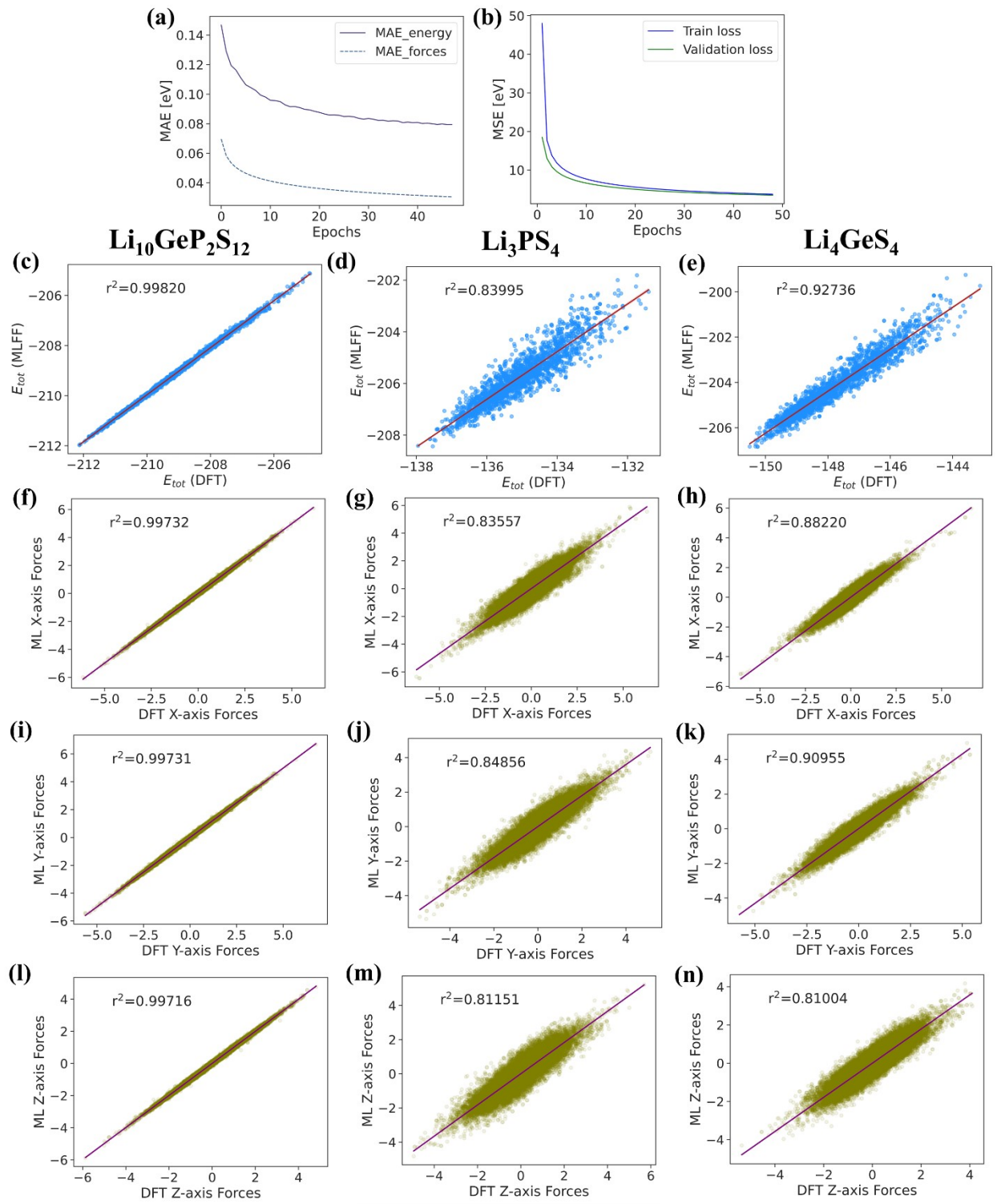

Figure Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of SchNet model against AIMD reference data for different materials

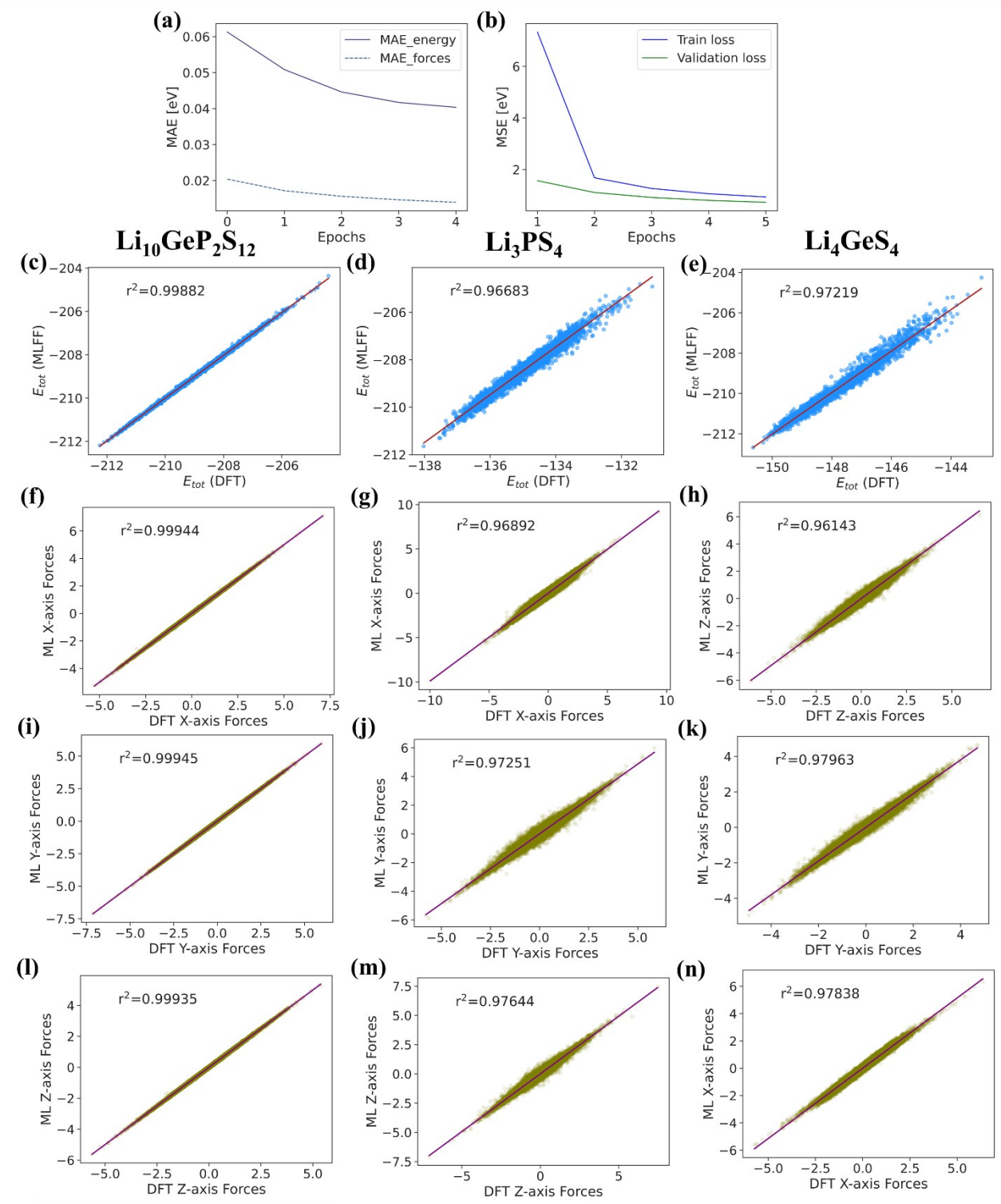

Figure S3 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of DimeNet model against AIMD reference data for different materials

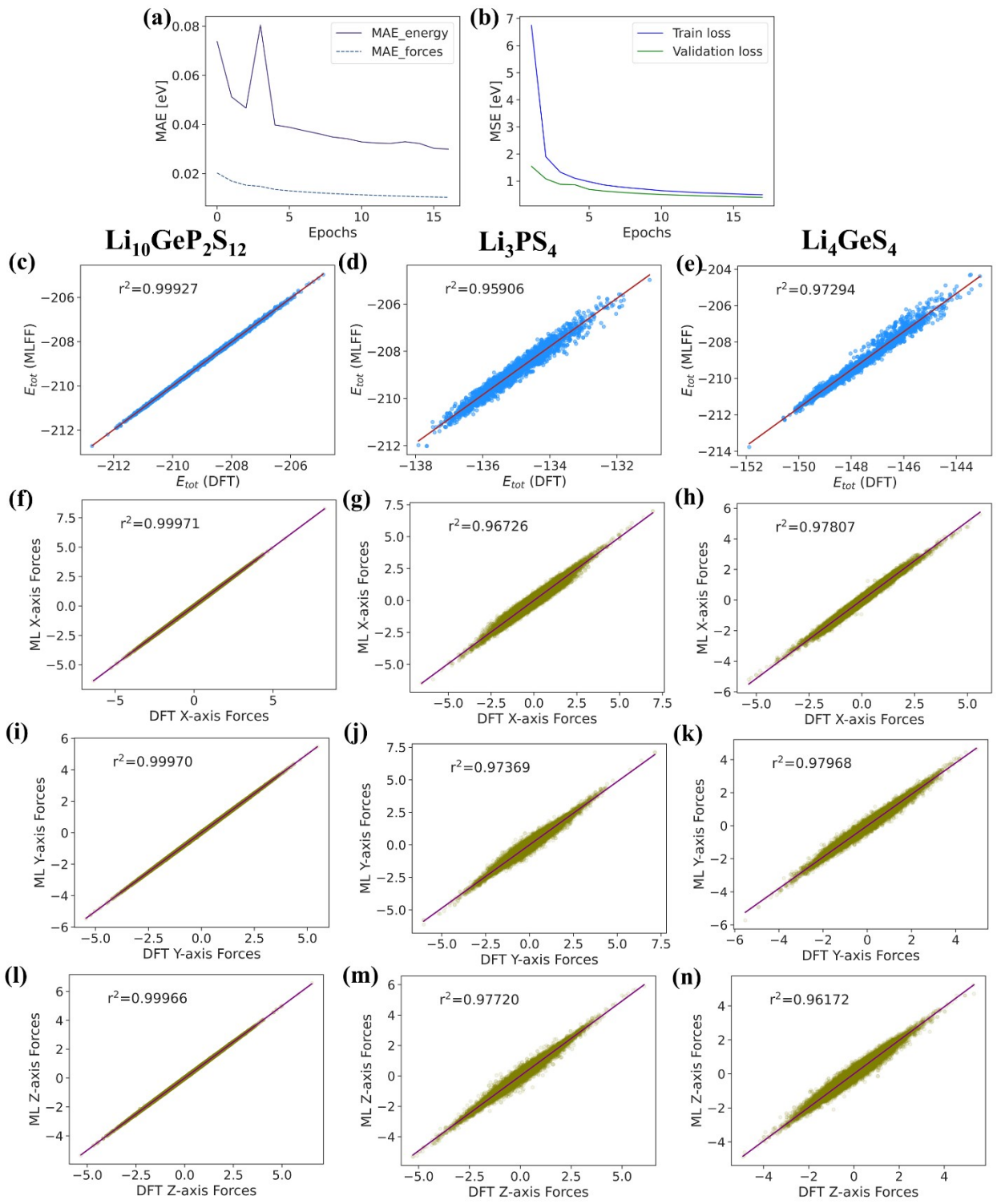

Figure S4 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of DimeNet++ model against AIMD reference data for different materials

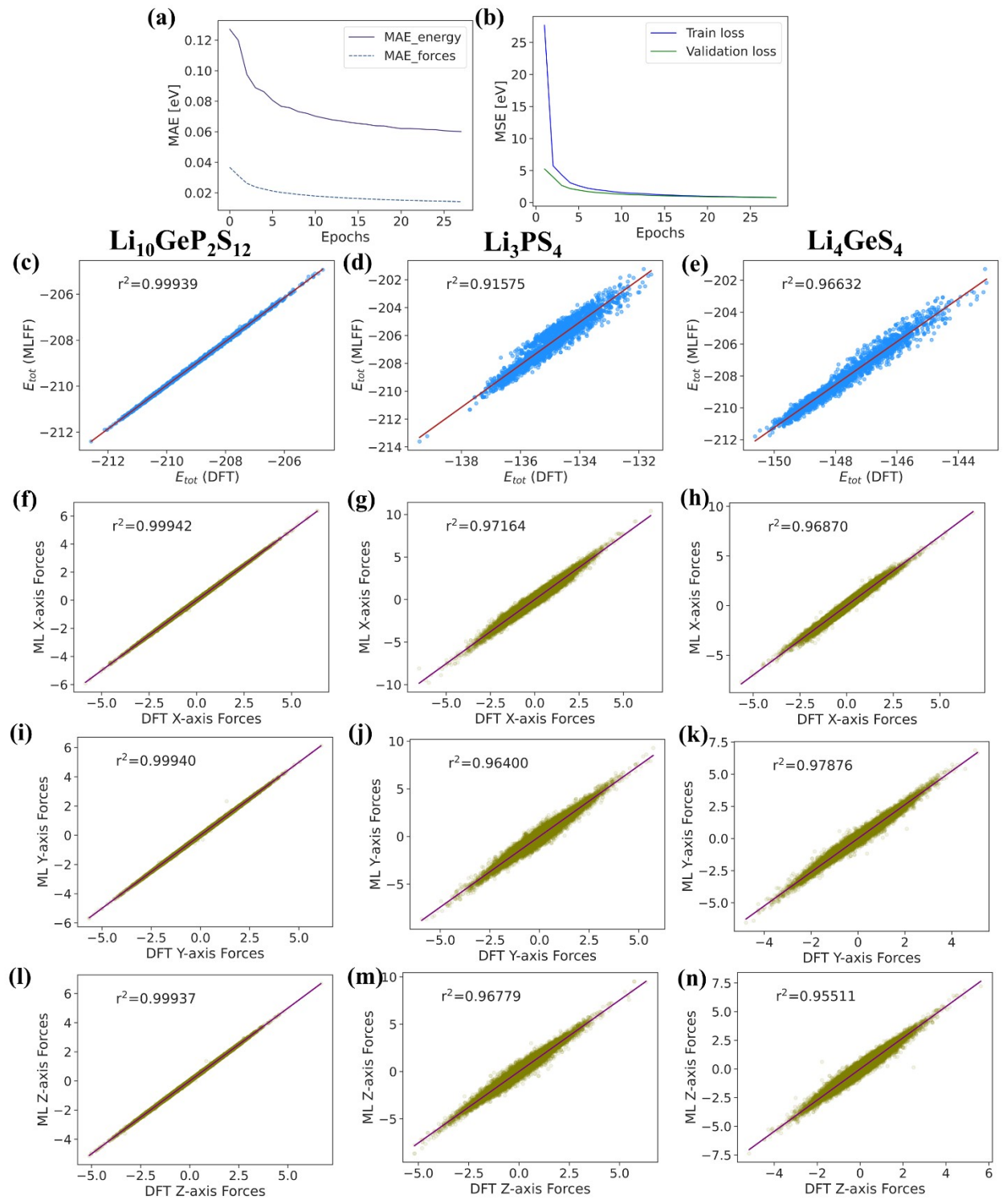

Figure S5 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of GNS-TAT model against AIMD reference data for different materials

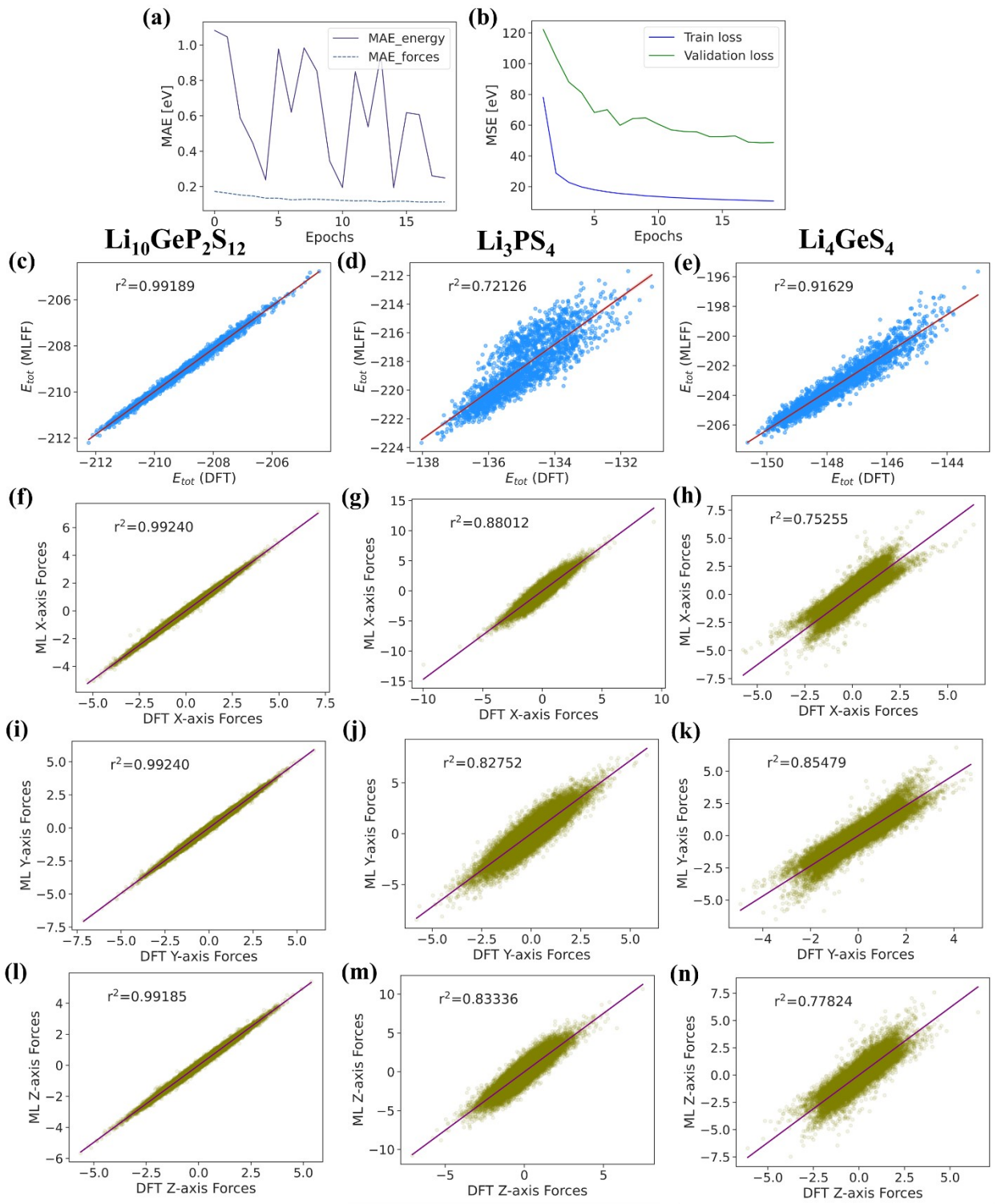

Figure S6 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of DeeperGATGNN model against AIMD reference data for different materials

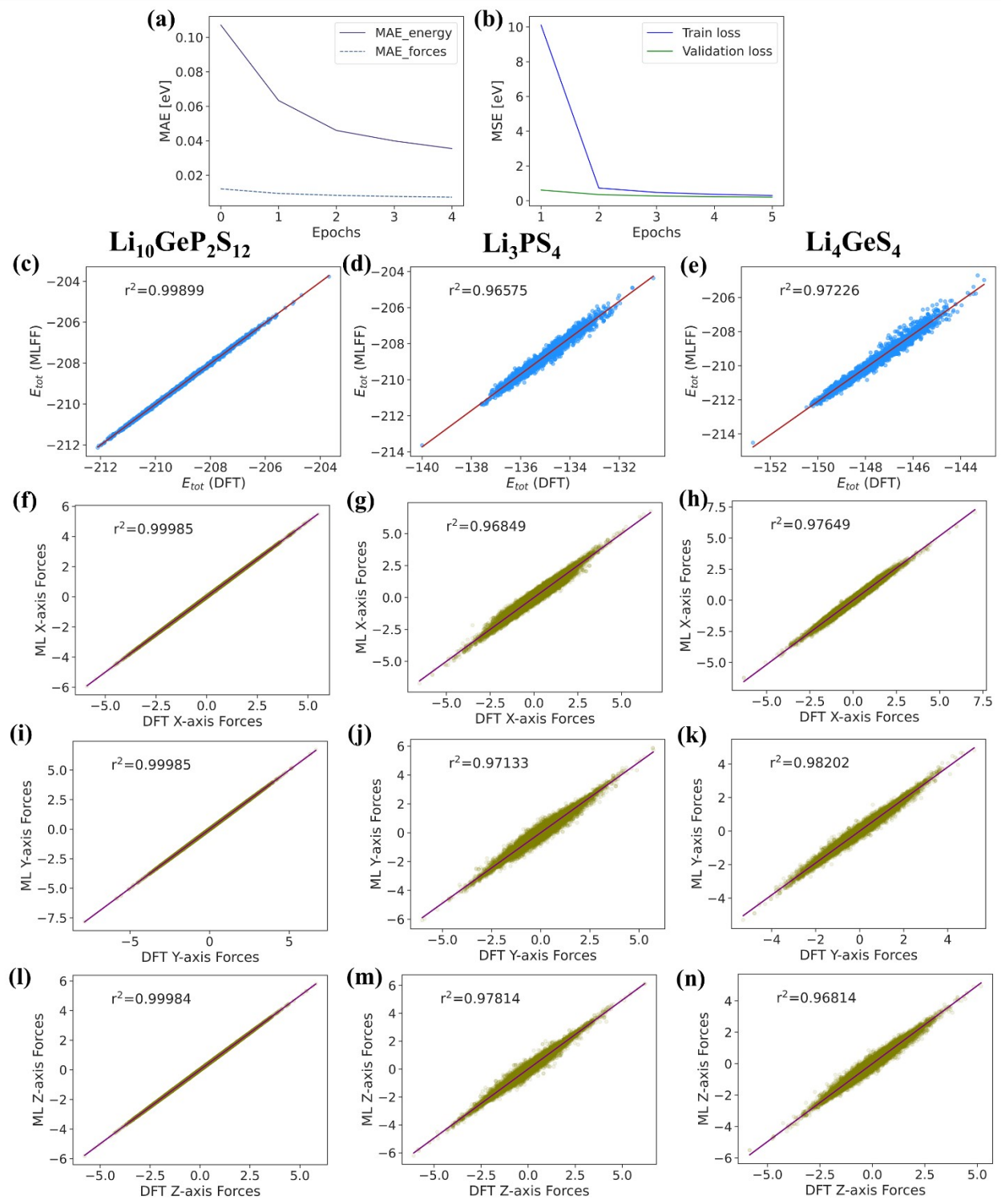

Figure S7 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of SCN model against AIMD reference data for different materials

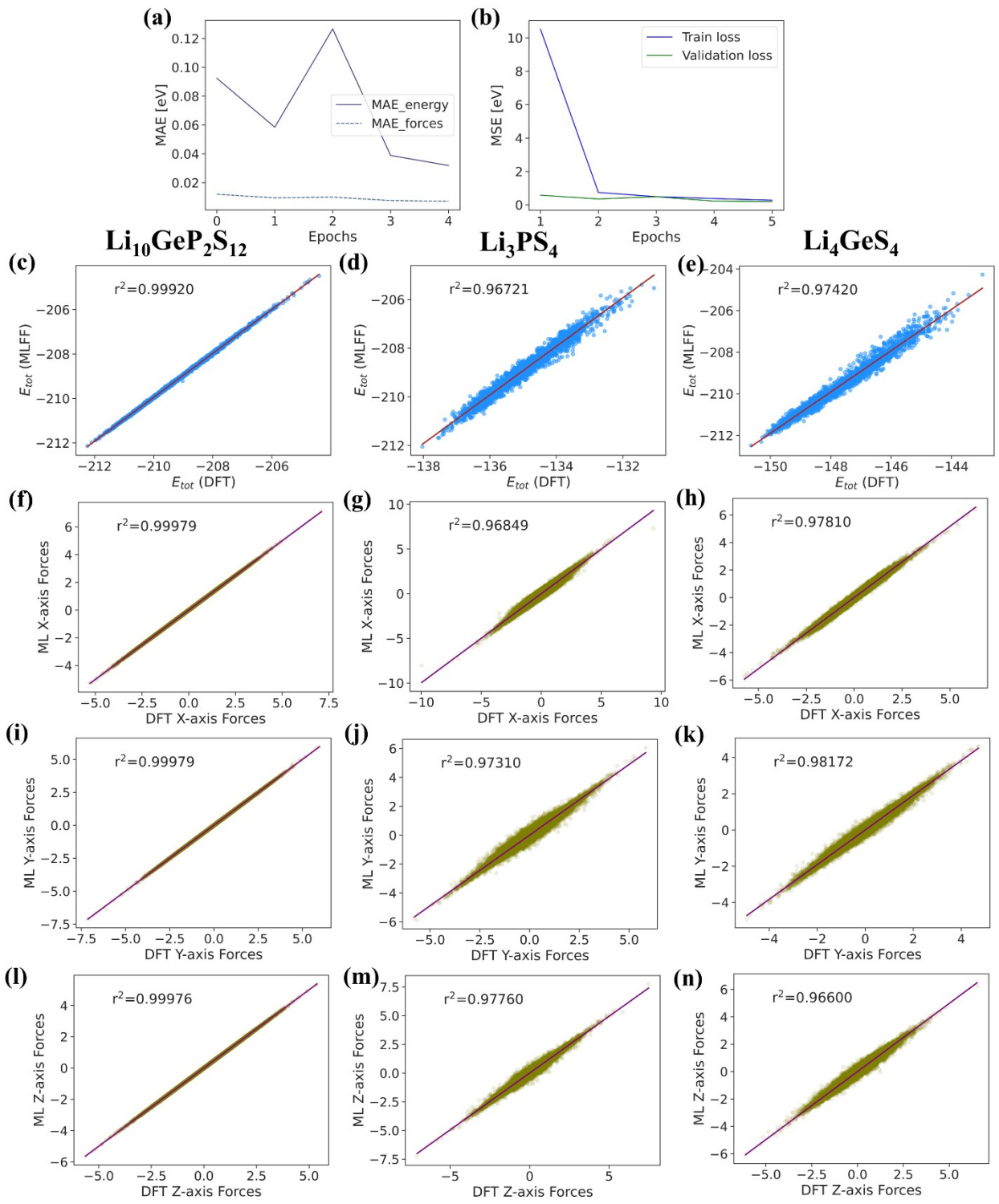

Figure S8 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of eSCN model against AIMD reference data for different materials

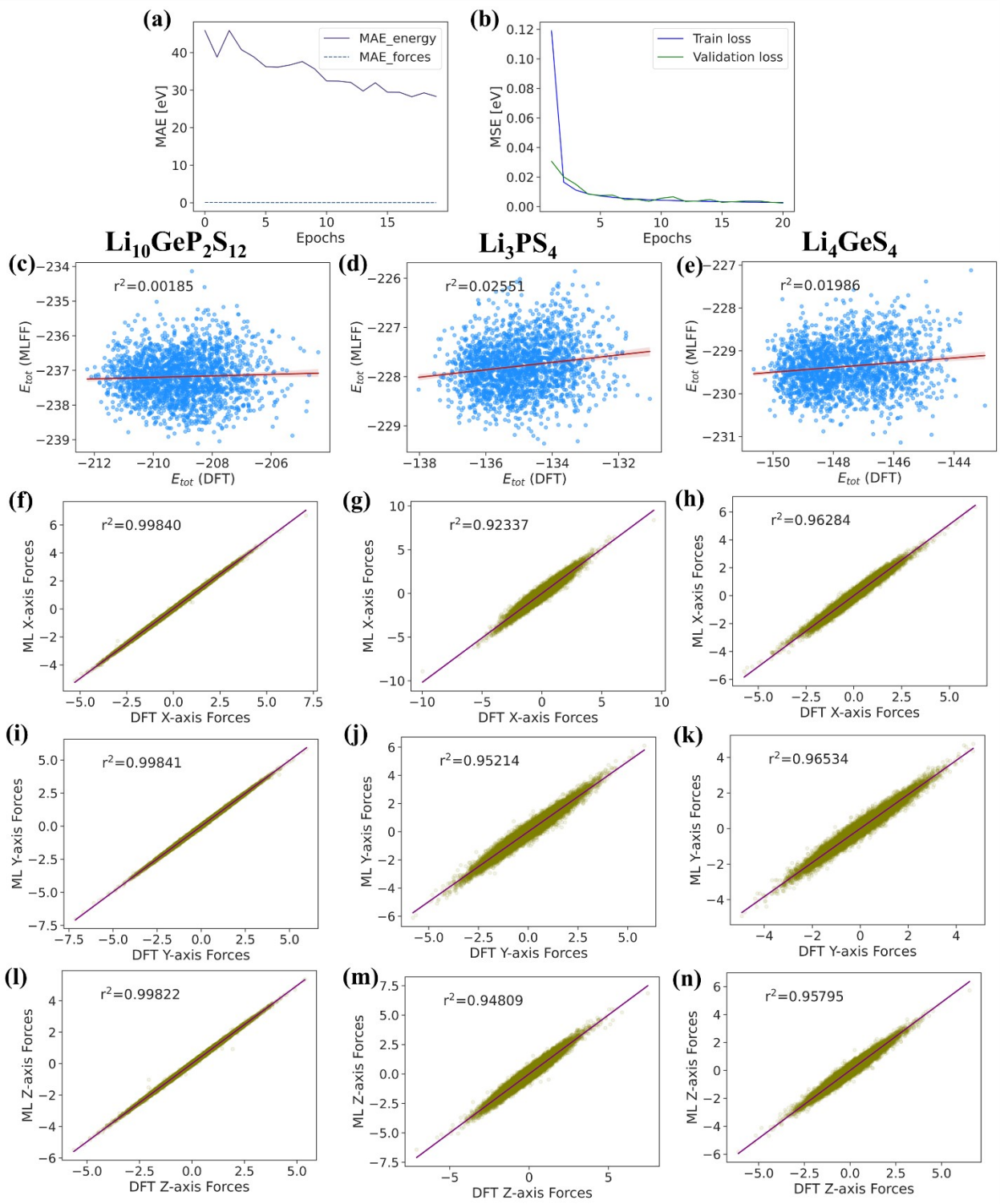

Figure S9 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of ForceNet model against AIMD reference data for different materials

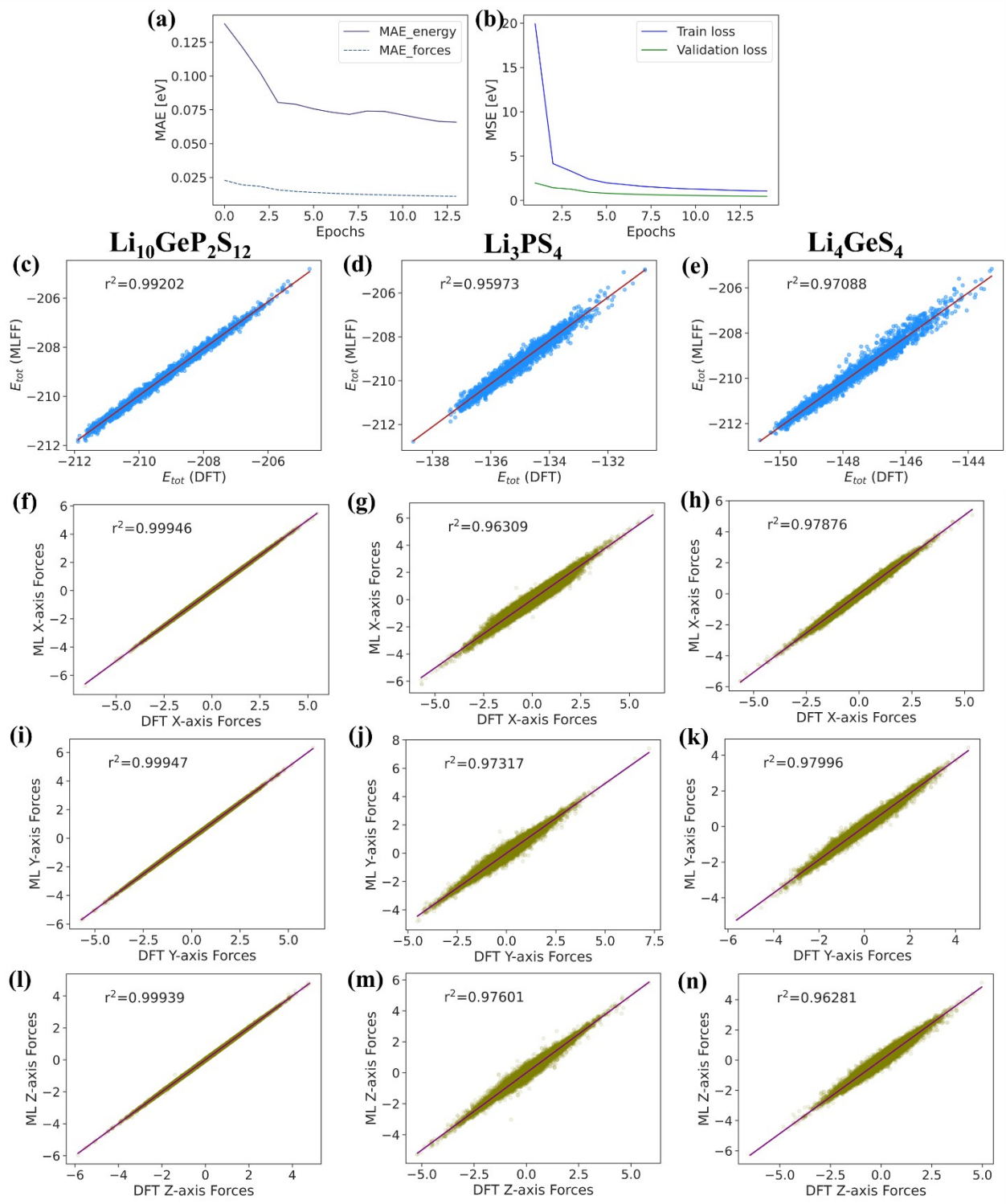

Figure S10 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of Equiformer model against AIMD reference data for different materials

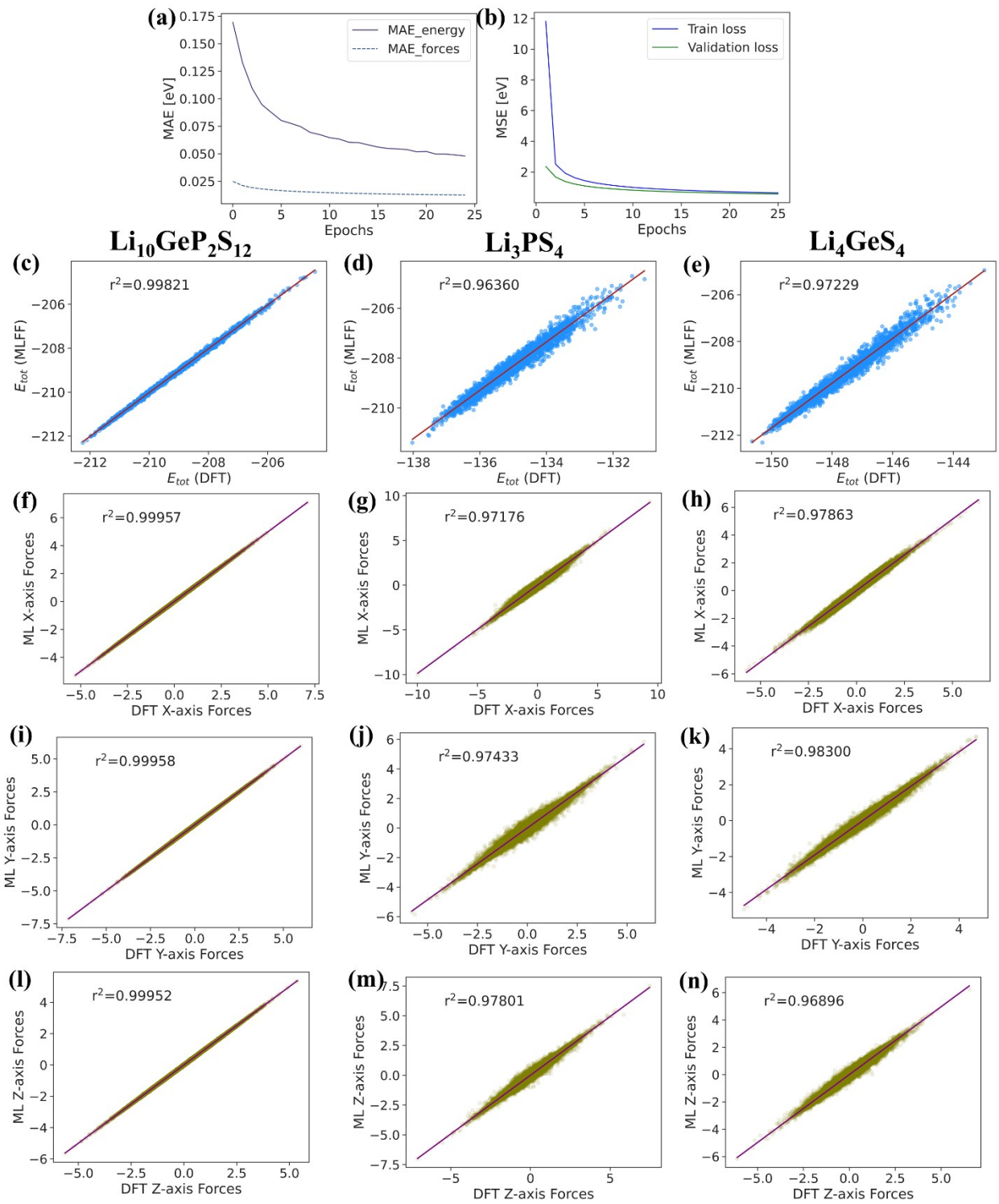

Figure S11 Training metrices (a) and losses (b) along with their direct energy and forces predictions (c–n) of LeftNet model against AIMD reference data for different materials

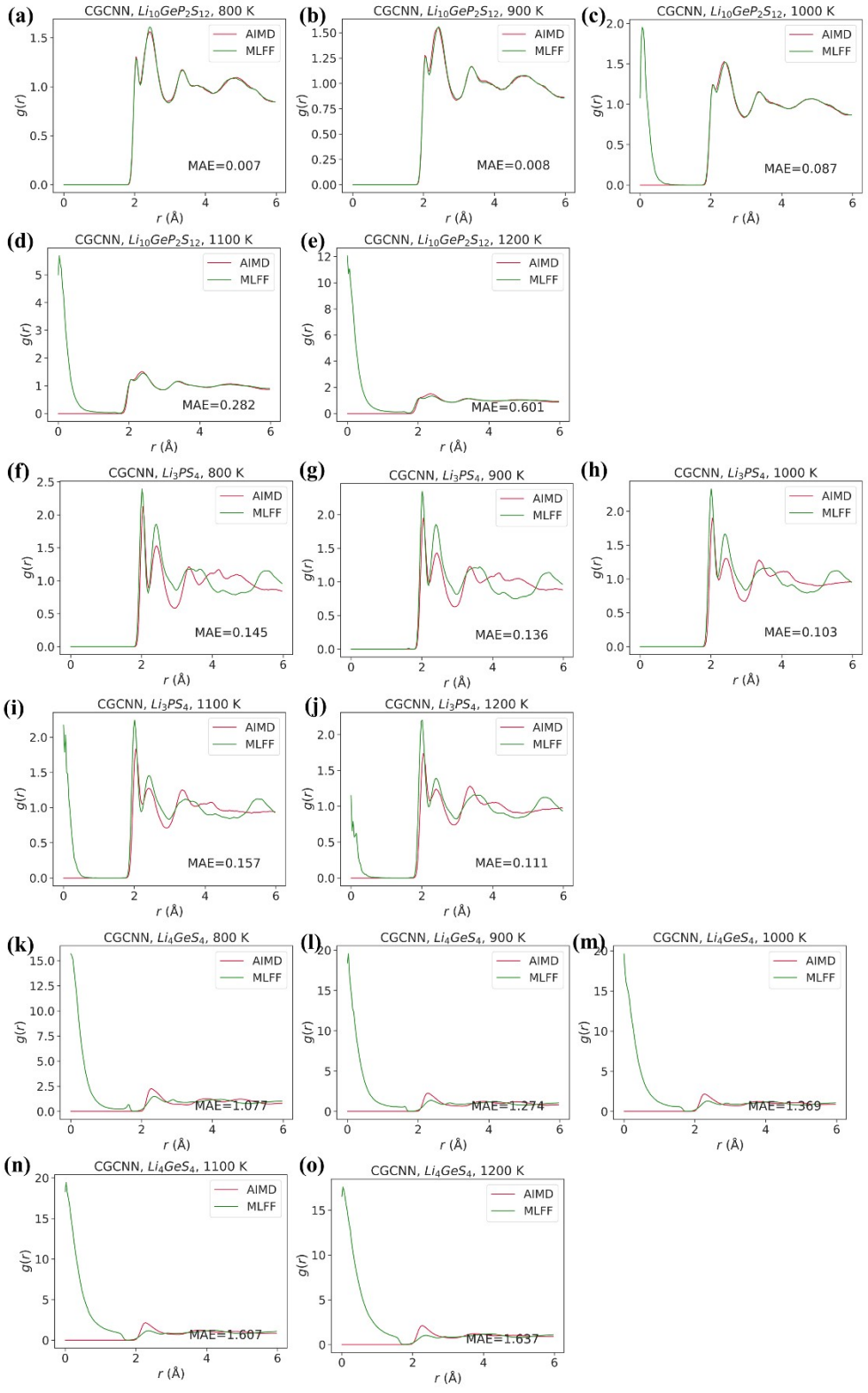

Figure S12 Radial distribution function prediction of CGCNN model against AIMD reference data for different materials

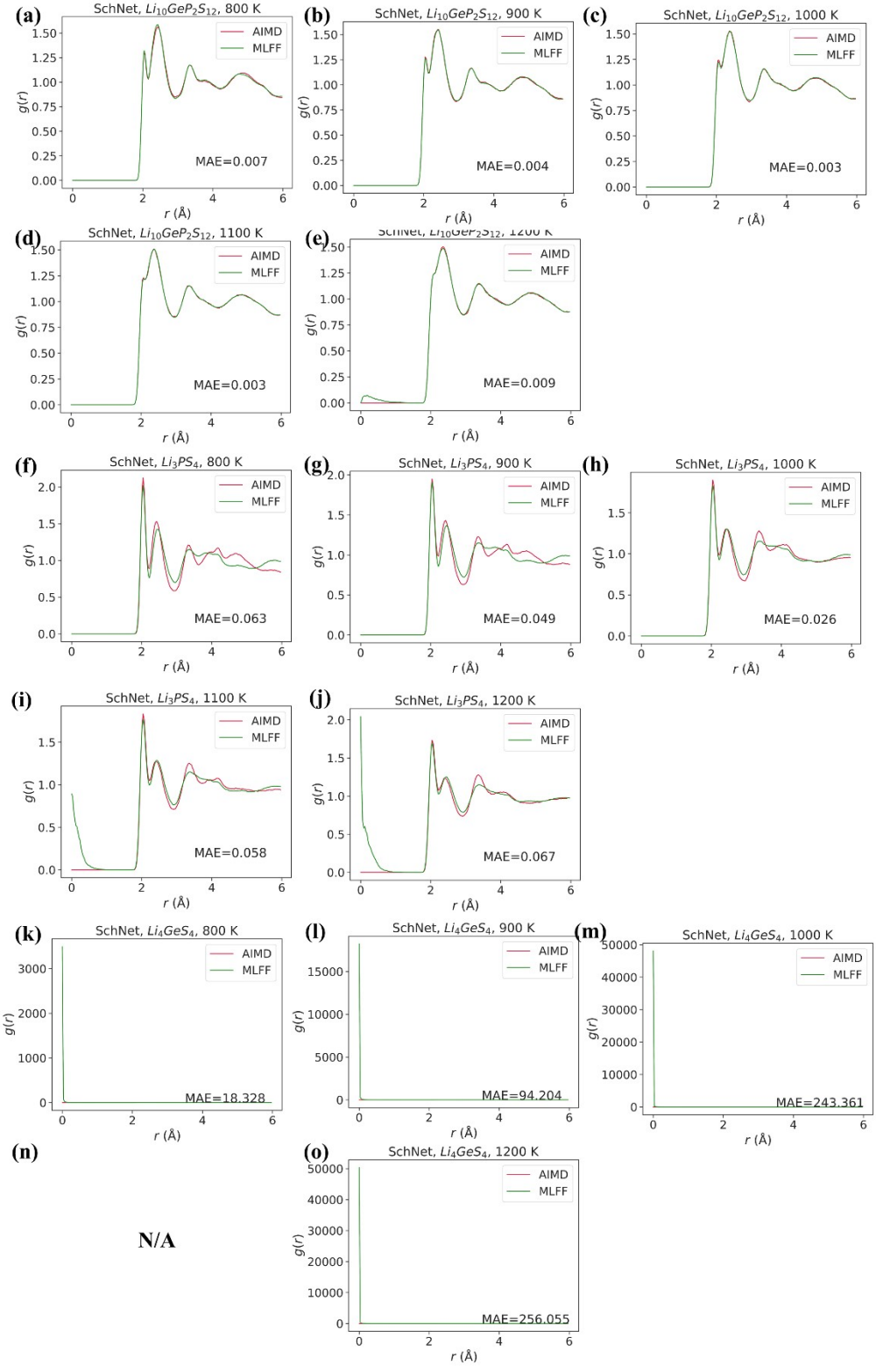

Figure S13 Radial distribution function prediction of SchNet model against AIMD reference data for different materials

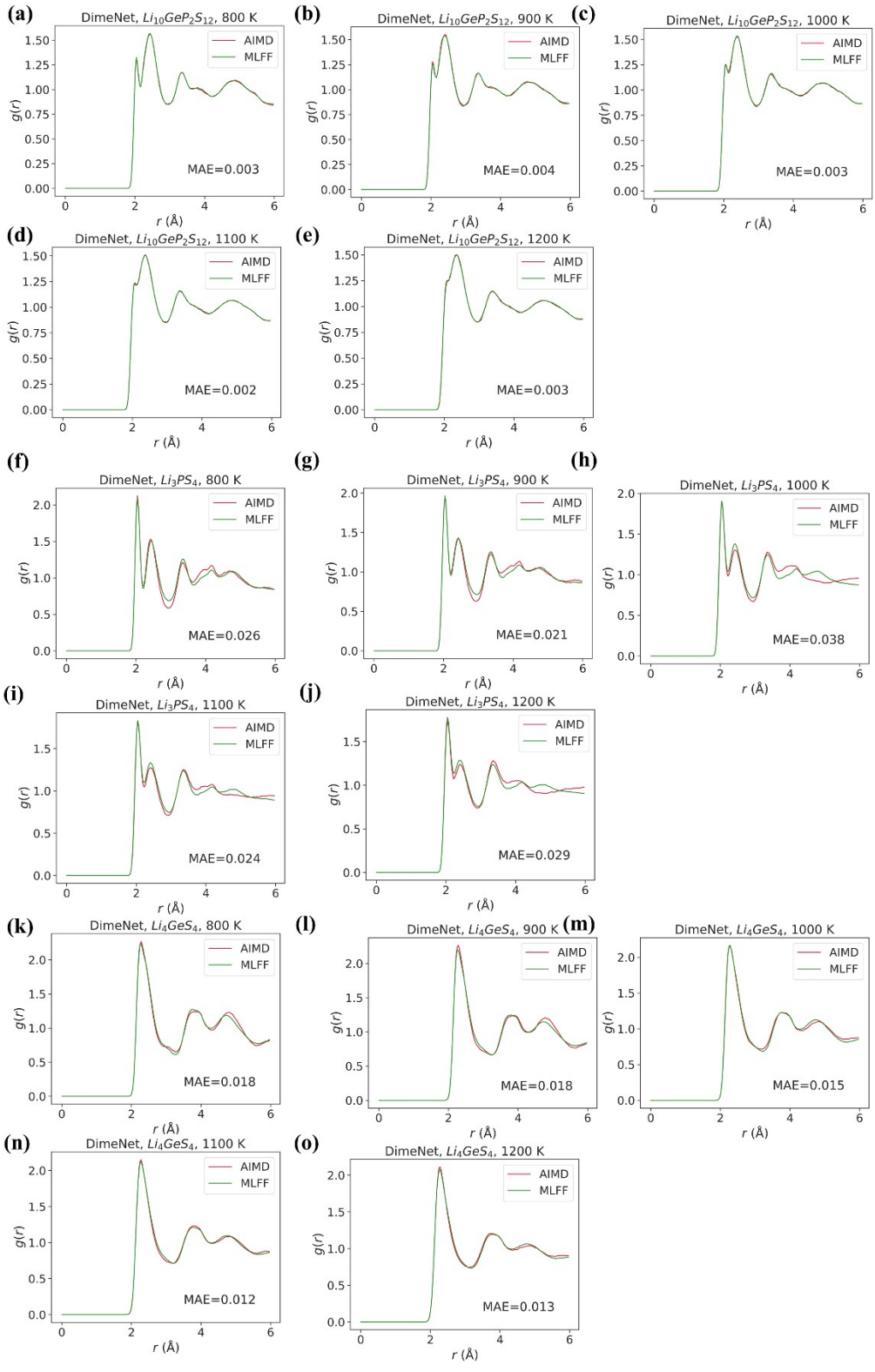

Figure S14 Radial distribution function prediction of DimeNet model against AIMD reference data for different materials

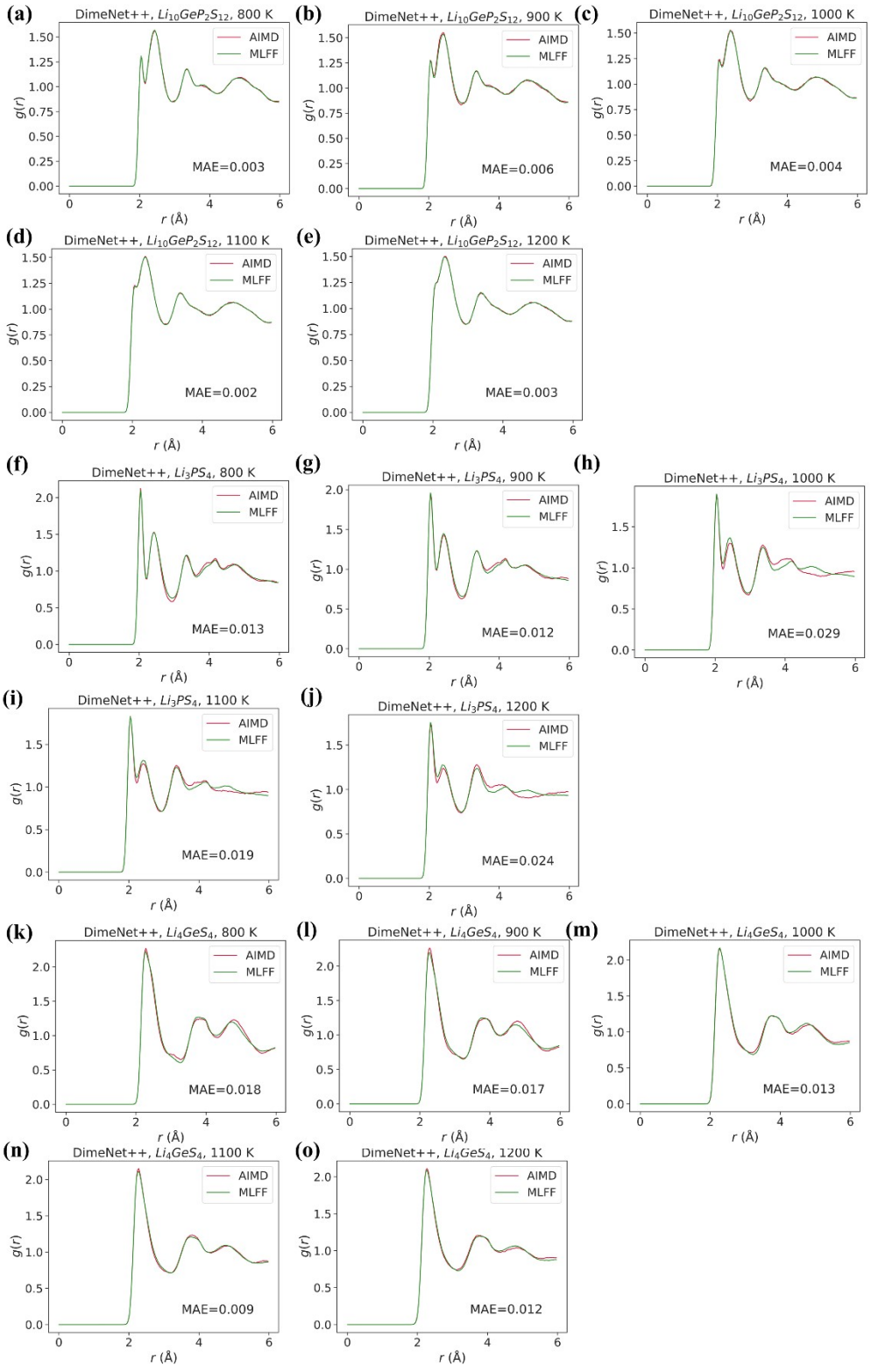

Figure S15 Radial distribution function prediction of DimeNet++ model against AIMD reference data for different materials

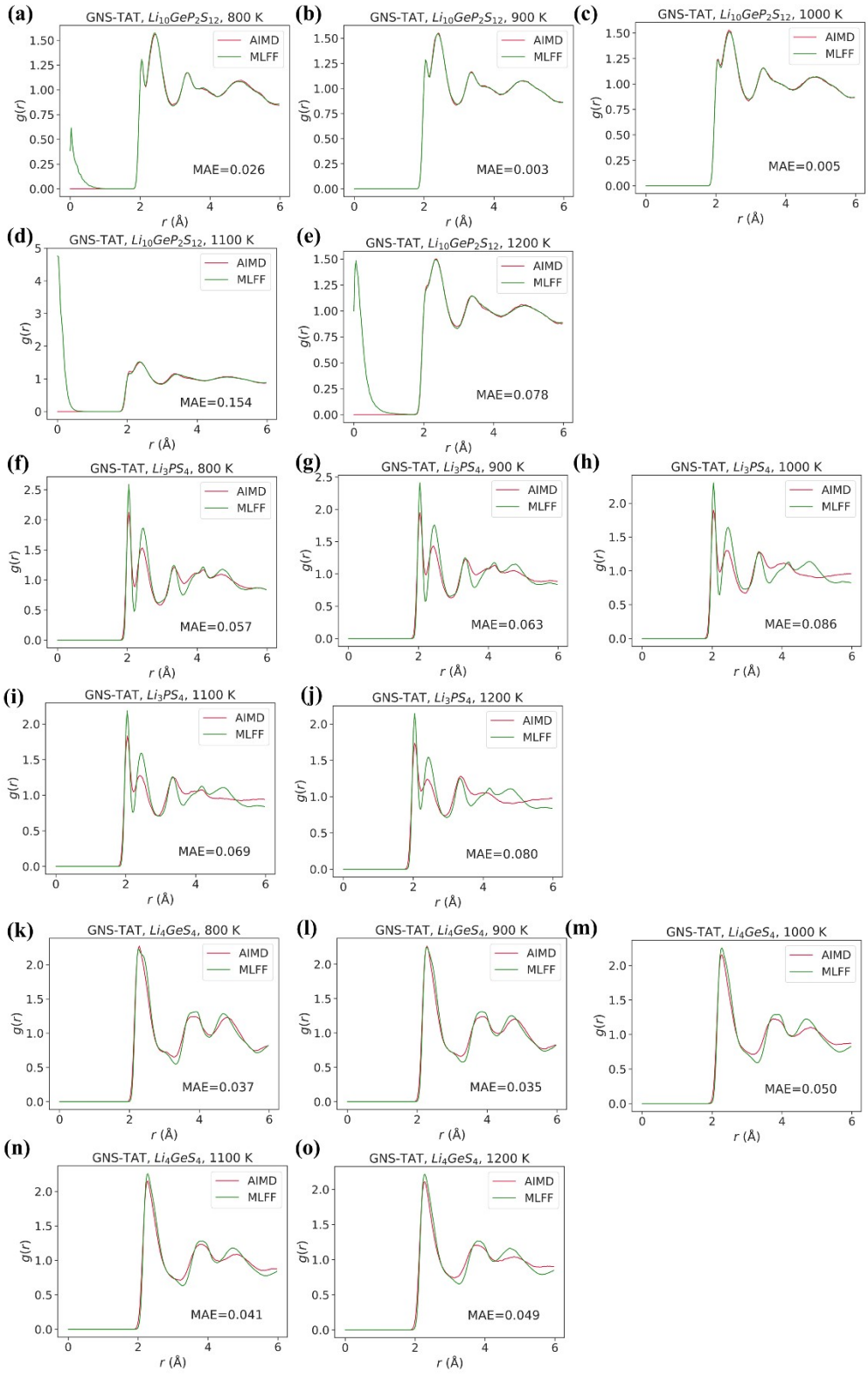

Figure S16 Radial distribution function prediction of GNS-TAT model against AIMD reference data for different materials

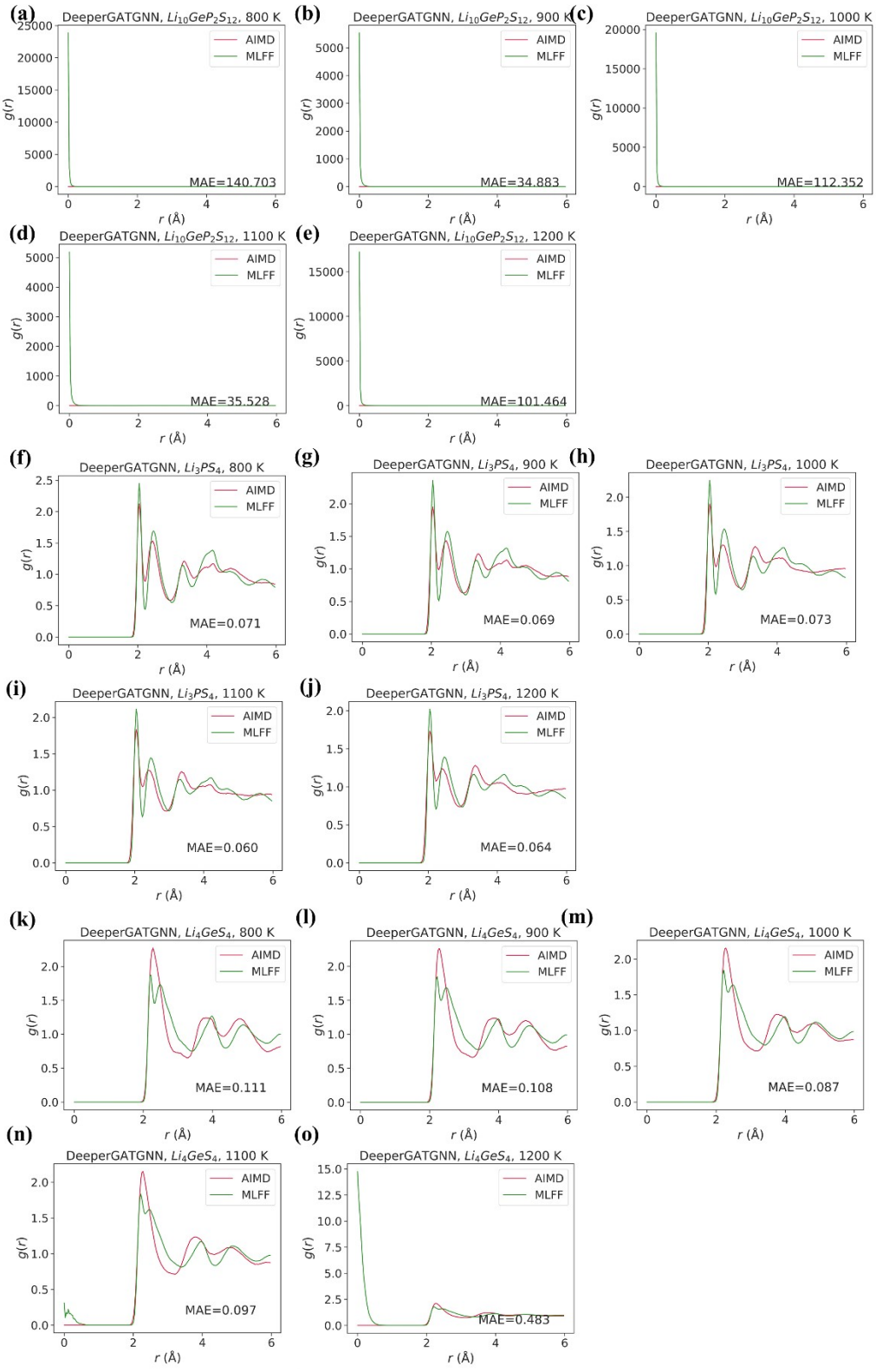

Figure S17 Radial distribution function prediction of DeeperGATGNN model against AIMD reference data for different materials

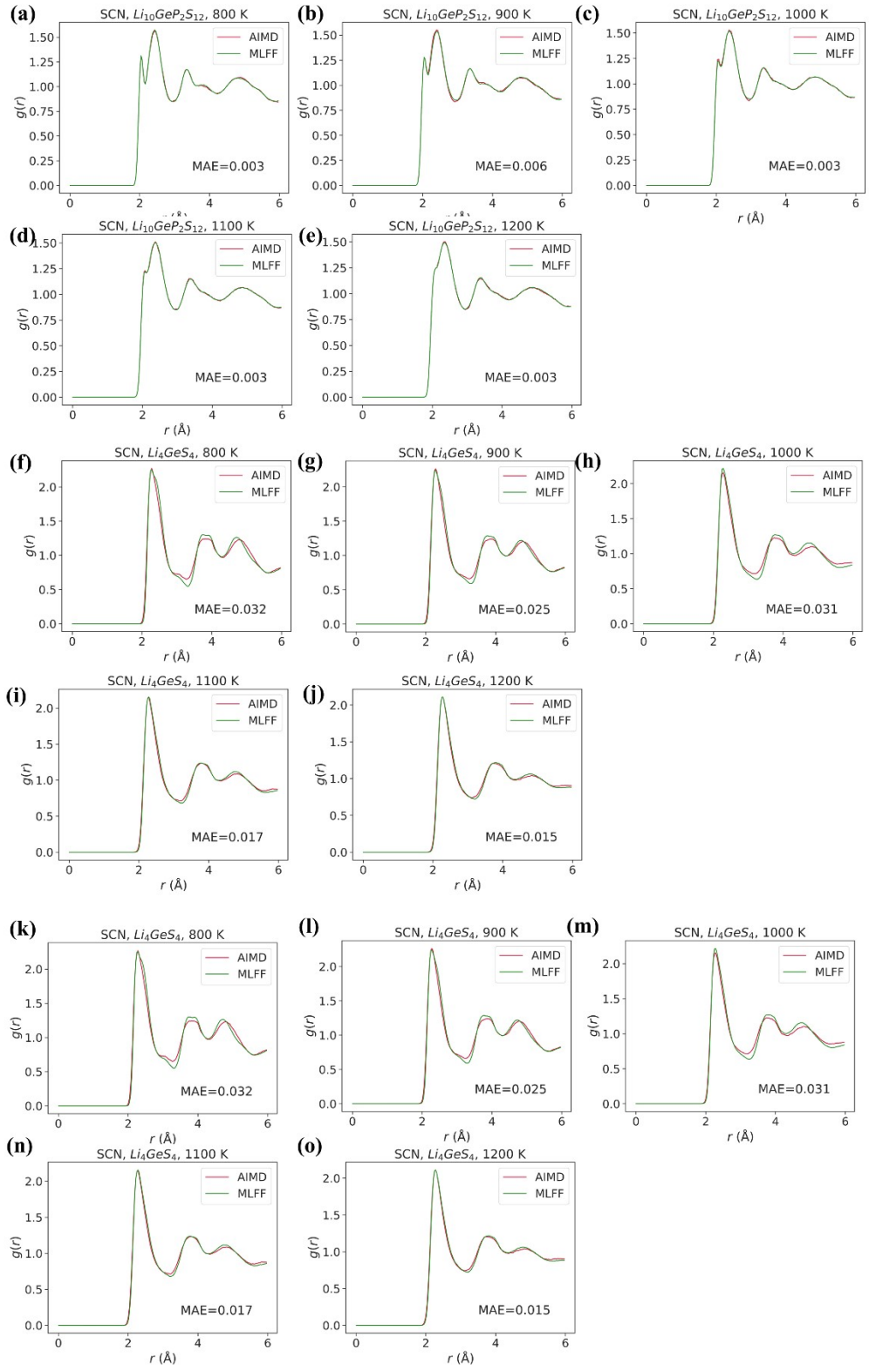

Figure S18 Radial distribution function prediction of SCN model against AIMD reference data for different materials

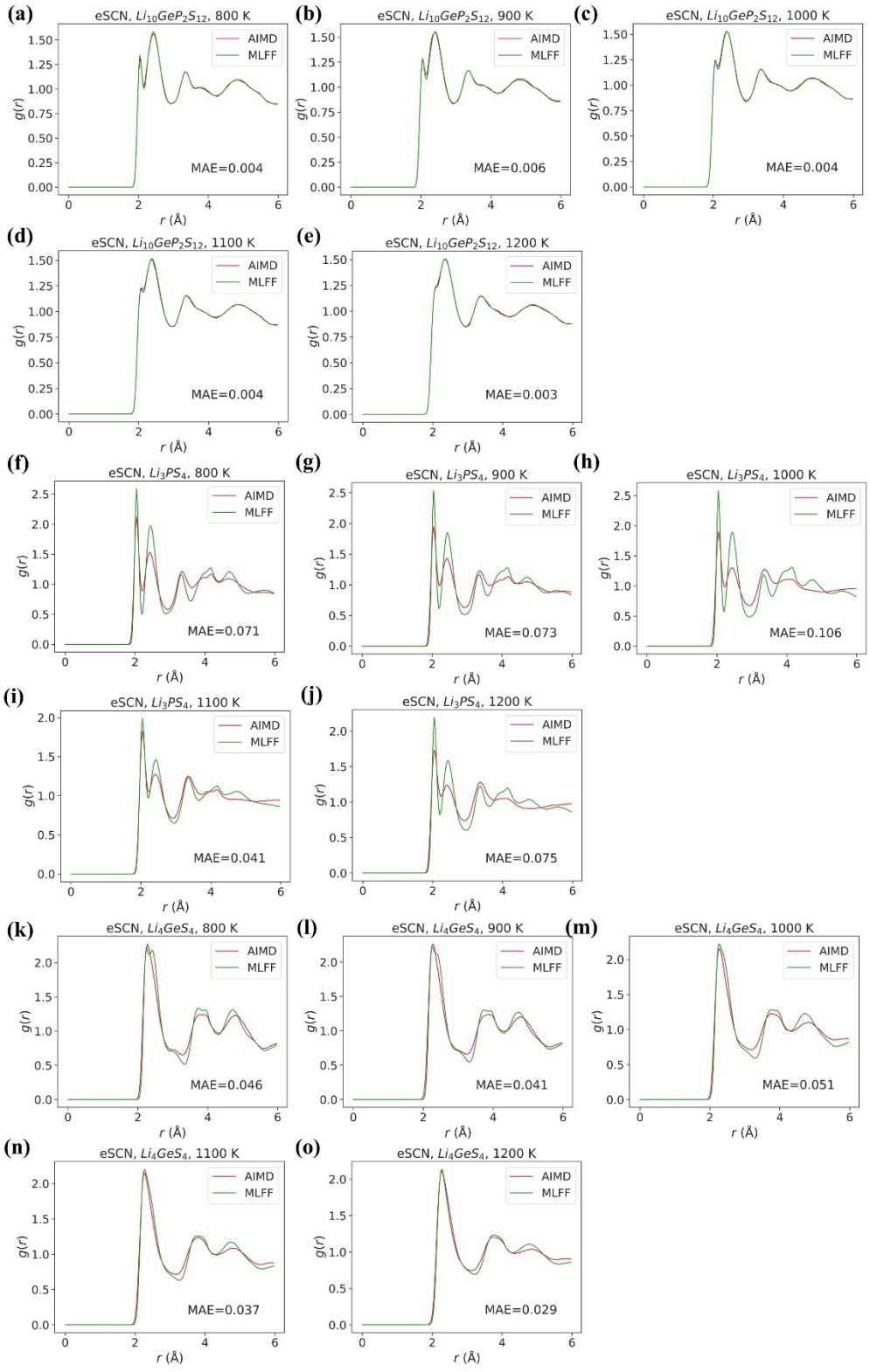

Figure S19 Radial distribution function prediction of eSCN model against AIMD reference data for different materials

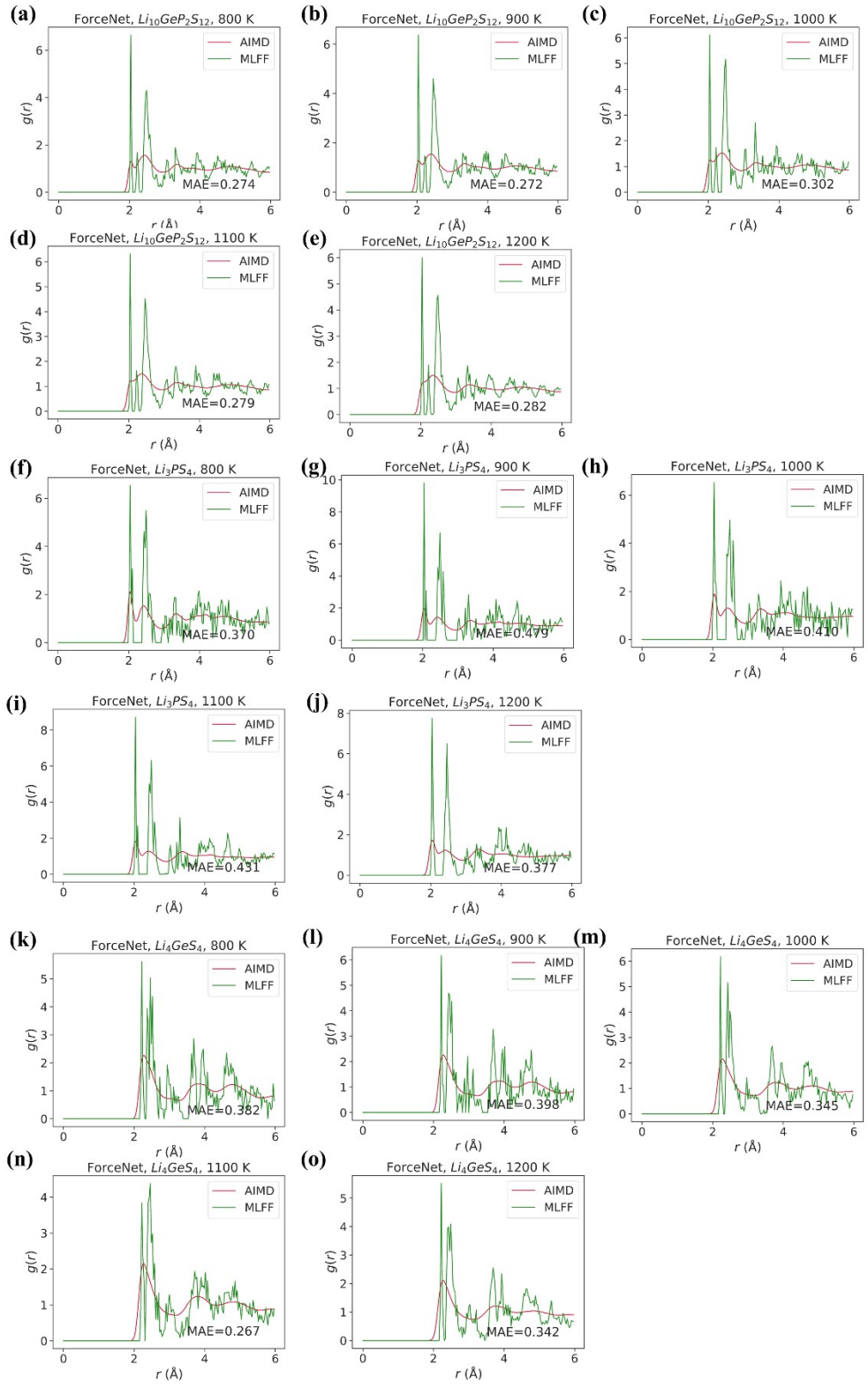

Figure S20 Radial distribution function prediction of ForceNet model against AIMD reference data for different materials

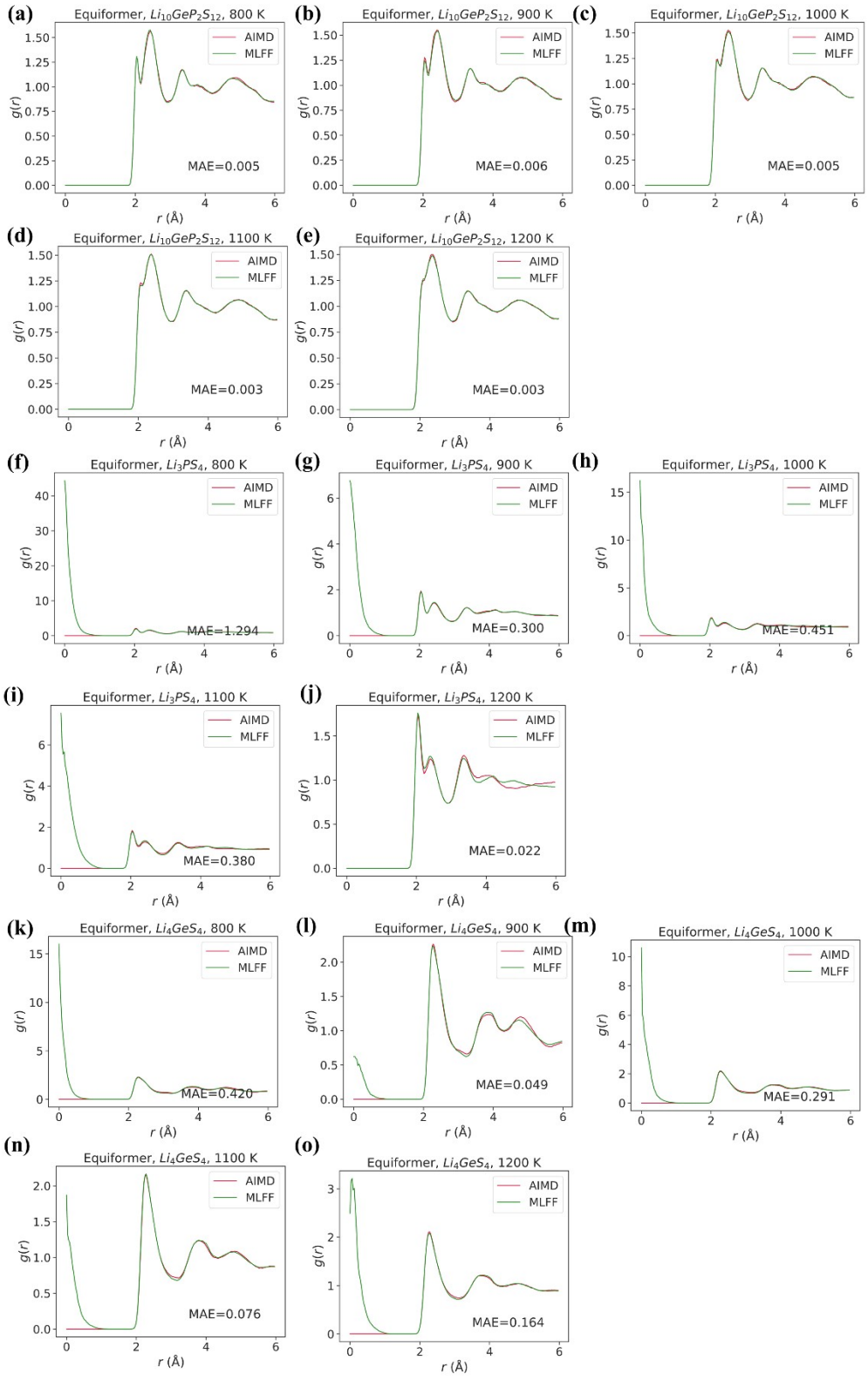

Figure S21 Radial distribution function prediction of Equiformer model against AIMD reference data for different materials

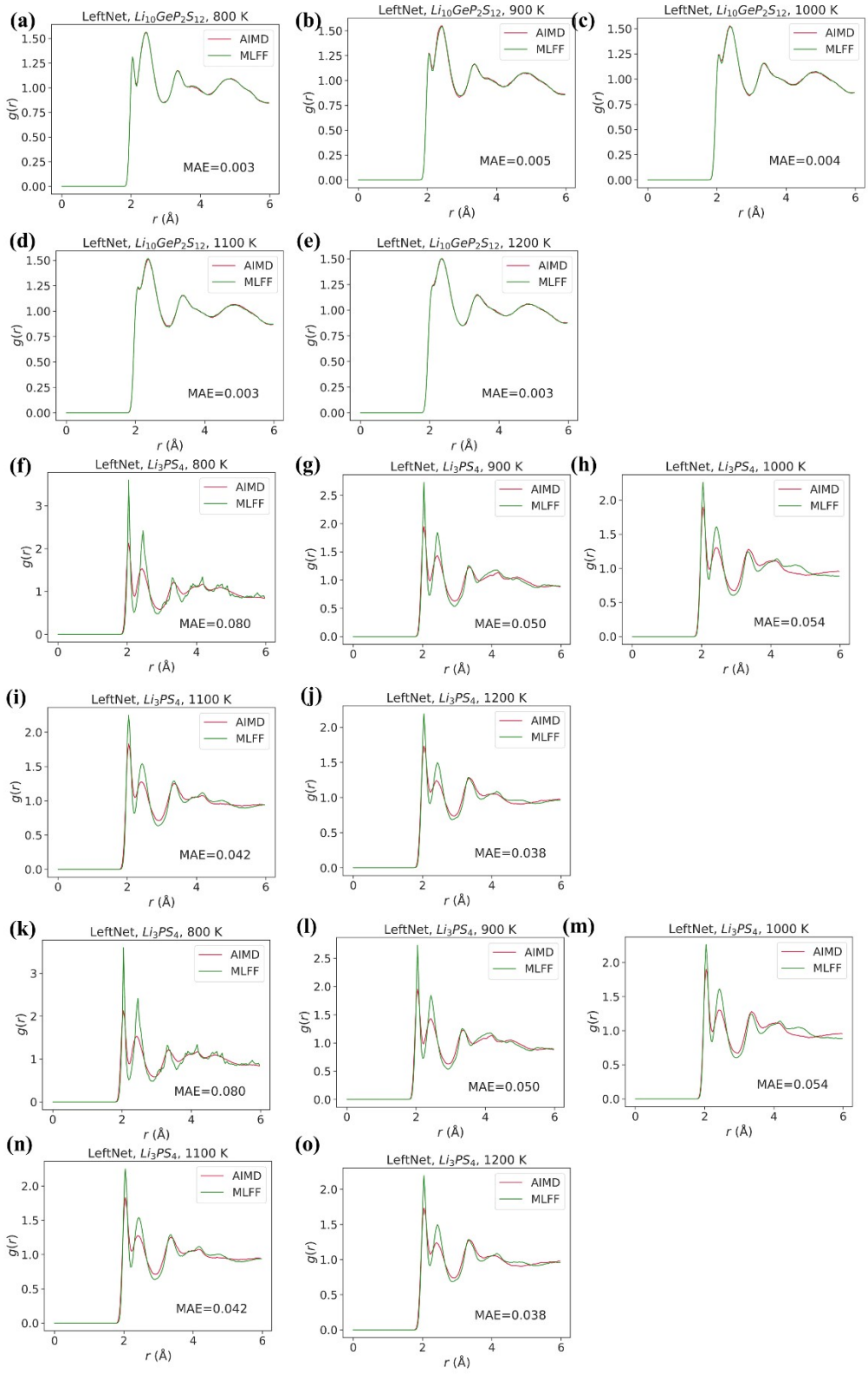

Figure S22 Radial distribution function prediction of LeftNet model against AIMD reference data for different materials

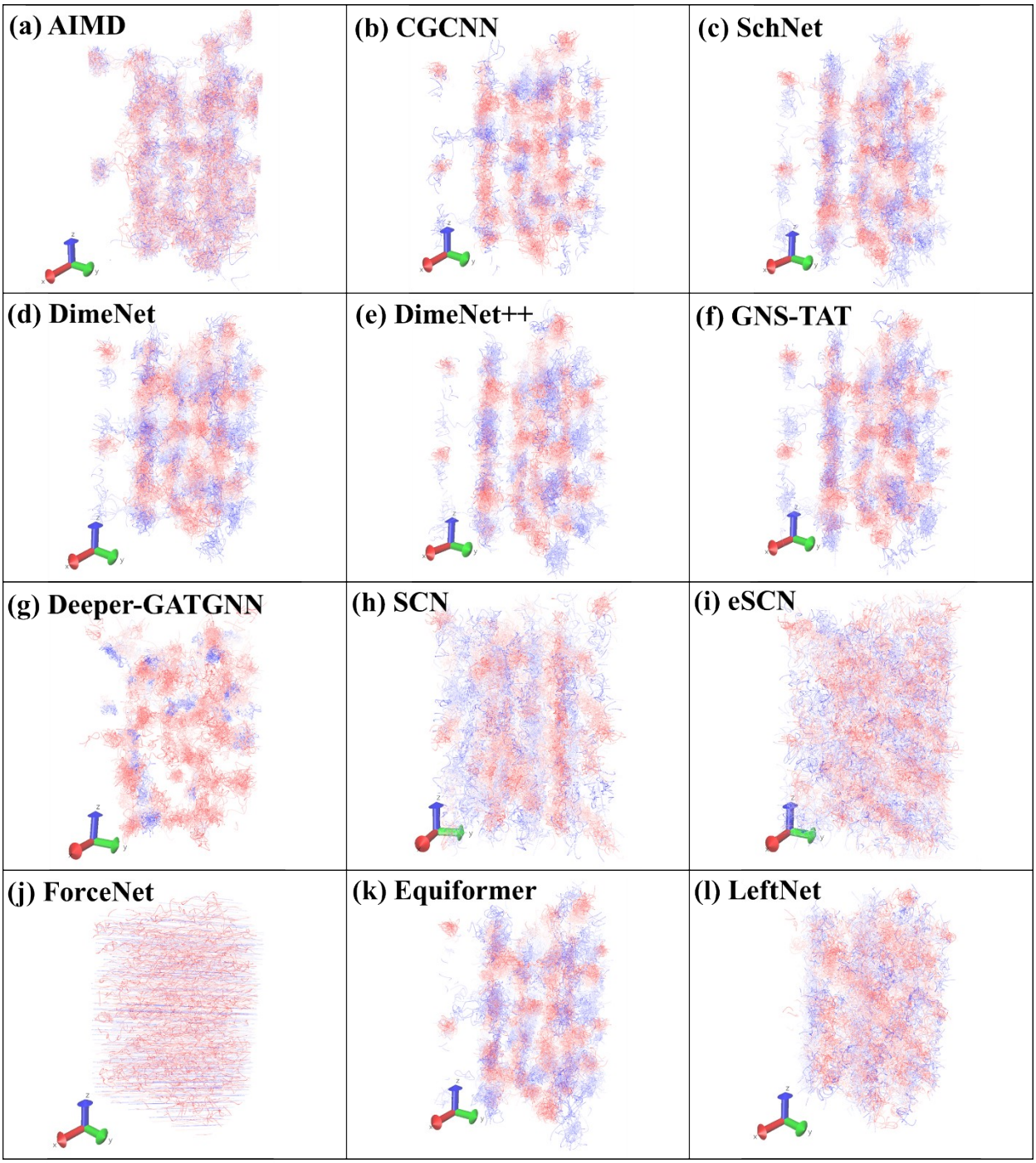

Figure S23 Time evolution of MLFF-MD trajectories for Li Atoms within the simulation system of $Li_{10}GeP_2S_{12}$ over 50 ps at 800K.

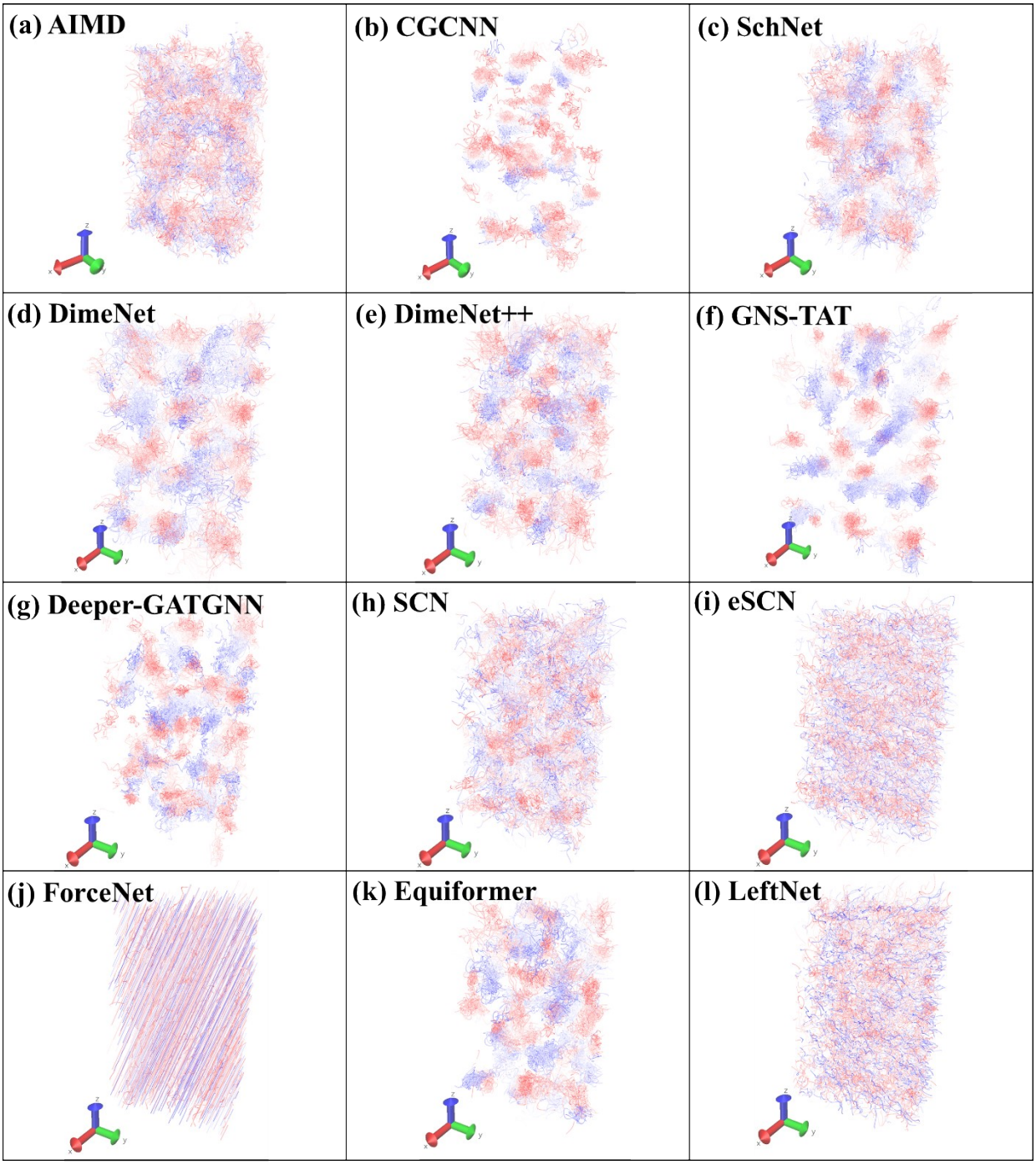

Figure S24 Time evolution of MLFF-MD trajectories for Li Atoms within the simulation system of Li_3PS_4 over 50 ps at 800K.

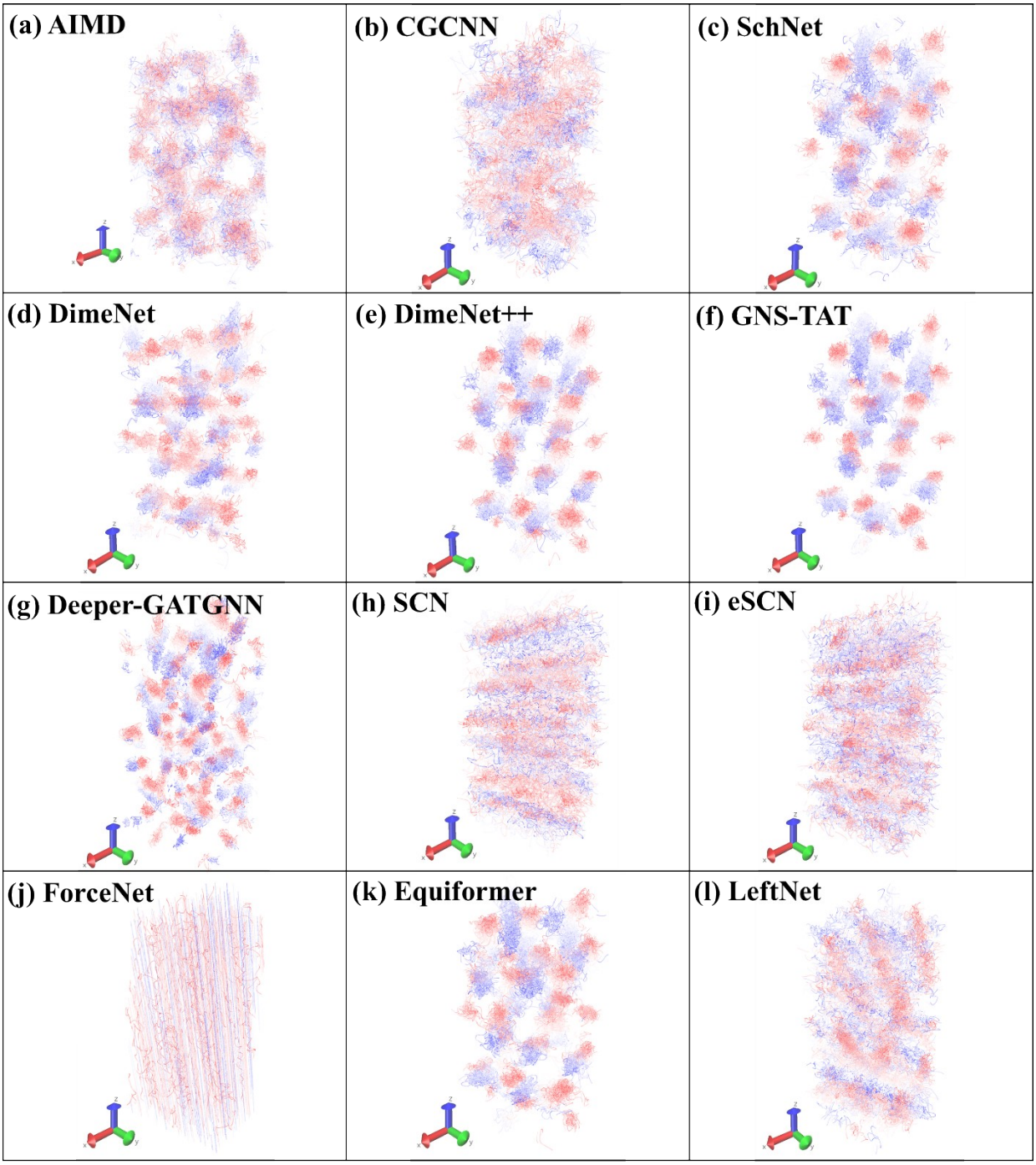

Figure S25 Time evolution of MLFF-MD trajectories for Li Atoms within the simulation system of Li_4GeS_4 over 50 ps at 800K.