Supplementary Information: A new universal force-field for the Li_2S–P_2S_5 system

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1 LAMMPS inputs

LAMMPS (3 Mar 2020) input files used for the validation are listed:

- γ-Li_3PS_4: gamma-Li3PS4.in gamma-Li3PS4.data
- β-Li_3PS_4: beta-Li3PS4.in beta-Li3PS4.data
- $Li_4P_2S_6$: Li4P2S6.in Li4P2S6.data
- $Li_7P_3S_{11}$: Li7P3S11.in Li7P3S11.data
- Li_7PS_6: Li7PS6.in Li7PS6.data
- 67Li_2S-33P_2S_5 glass: 67Li2S-33P2S5.in 67Li2S-33P2S5.data
- 70Li_2S-30P_2S_5 glass: 70Li2S-30P2S5.in 70Li2S-30P2S5.data
- 75Li_2S-25P_2S_5 glass: 75Li2S-25P2S5.in 75Li2S-25P2S5.data
- Three-body potential: Li-S.sw

2 Force-field parameters

Table S1: Lennard-Jones pair parameters of atoms i and j in Class II potential. Parameters for a combination of different two types of P2, Pa, P4, S2, S1, Sc, or S4 are derived from Waldman–Hagler combining rules.

i	j	ε/kJ/mol	r^0/Å
Li	Li	0	0
Li	P2	0	0
Li	Pa	0	0
Li	P4	0	0
Li	S2	3.18402	2.87451
Li	S1	6.87732	2.68218
Li	Sc	5.70836	2.65312
Li	S4	4.56893	2.73400
P2	P2	102.307	0.935300
P2	Pa	85.2078	0.845927
P2	P4	88.9600	1.28948
P2	S2	0.320577	3.62767
P2	S1	0.111702	4.10206
P2	Sc	0.805645	3.13883
P2	S4	0.0406344	4.70108
Pa	Pa	224.376	0.631531
Pa	P4	43.4183	1.27491
Pa	S2	0.146170	3.62759
Pa	S1	0.0509280	4.10201
Pa	Sc	0.367408	3.13866
Pa	S4	0.0185256	4.70106
P4	P4	286.462	1.42927
P4	S2	1.91098	3.62871
P4	S1	0.666463	4.10262
P4	Sc	4.79110	3.14097
P4	S4	0.242553	4.70137
S2	S2	1.71023	4.07182
S2	S1	0.806141	4.37816
S2	Sc	2.54094	3.84569
S2	S4	0.357938	4.85356
S1	S1	0.434036	4.60435
S1	Sc	1.04667	4.22896
S1	S4	0.219075	4.99641
Sc	Sc	4.53320	3.52301
Sc	S4	0.419943	4.76802
S4	S4	0.130121	5.27676
Li	S0	4.86227	2.83510
P4	S0	2.76741	4.01127
S4	S0	0.826743	4.96363
S0	S0	6.53884	4.50173

Table S2: Bond parameters of bonded atoms i-j in Class II potential.

i	j	r_0/Å	K_2/kJ/mol/Å	K_3/kJ/mol/Å^3	K_4/kJ/mol/Å^4
P2	S2	2.05800	759.383	243.108	168.519
Pa	S1	2.06993	673.925	90.4907	2326.70
Pa	Sc	2.28618	327.446	78.7211	1192.39
P4	S4	2.09744	508.620	26.5935	2697.60
P2	P2	2.31455	659.411	627.968	601.387

Table S3: Angle parameters of bonded atoms i-j-k with j center in Class II potential.

i	j	k	θ_0/°	K_2/kj/mol/rad^2	K_3/kj/mol/rad^3	K_4/kj/mol/rad^4
P2	P2	S2	104.305	135.767	203.550	769.375
Pa	Sc	Pa	116.460	355.808	45.8048	1984.85
S2	P2	S2	115.313	177.728	267.296	1806.27
S1	Pa	S1	114.173	216.016	48.6377	191.055
S1	Pa	Sc	105.522	149.132	38.1655	416.925
S4	P4	S4	109.460	201.806	75.1756	803.675

Table S4: Bond-bond (bb) and bond-angle (ba) parameters of bonded atoms *i*-*j*-*k* with *j* center in Class II potential.

i	*j*	*k*	M/kJ/mol/Å	N_1/kJ/mol/Å	N_2/kJ/mol/Å^2	r_1/Å	r_2/Å
P2	P2	S2	21.5633	19.6593	111.905	2.26144	2.05203
Pa	Sc	Pa	397.935	211.374	249.228	2.09524	2.12546
S2	P2	S2	43.6646	183.480	224.394	1.96742	2.07272
S1	Pa	S1	89.8430	89.2376	84.0482	2.06174	2.02696
S1	Pa	Sc	47.7348	74.6585	103.964	2.07712	2.13569
S4	P4	S4	127.641	70.7761	96.2362	2.17645	2.18138

Table S5: Dihedral parameters of bonded atoms *i*-*j*-*k*-*l* in Class II potential.

i	*j*	*k*	*l*	K_1/kJ/mol	ϕ_1/°	K_2/kJ/mol	ϕ_2/°	K_3/kJ/mol	ϕ_3/°
Pa	Sc	Pa	S1	2.67041	195.783	3.16898	103.391	1.91883	133.865
S2	P2	P2	S2	0.0	0.0	0.0	0.0	0.0	0.0

Table S6: λ and $\cos\theta^0$ parameters with *i* center of nonbonded atoms *i*-*j*-*k* in Stillinger–Weber potential.

i	*j*	*k*	λ/kJ/mol	$\cos\theta^0$
Li	S2	S2	72.3481	−0.106294
Li	S2	S1	272.660	−0.276177
Li	S2	Sc	2151.02	−0.376578
Li	S2	S4	534.782	−0.317077
Li	S1	S1	797.550	−0.0672886
Li	S1	Sc	71.4410	−0.0212434
Li	S1	S4	217.683	−0.409570
Li	Sc	Sc	1438.18	0.124328
Li	Sc	S4	443.881	0.0118708
Li	S4	S4	50.7193	−0.250519
Li	S4	S0	33.3340	0.0535239
Li	S0	S0	2248.17	−0.348738

Table S7: γ parameters in Stillinger–Weber potential. $r^{\mathrm{cut}} = 5.0$ Å for all pairs.

i	*j*	γ /unitless
Li	S2	4.61683
Li	S1	5.47149
Li	Sc	5.11593
Li	S4	5.42257
Li	S0	5.26419

3 Mean square displacements

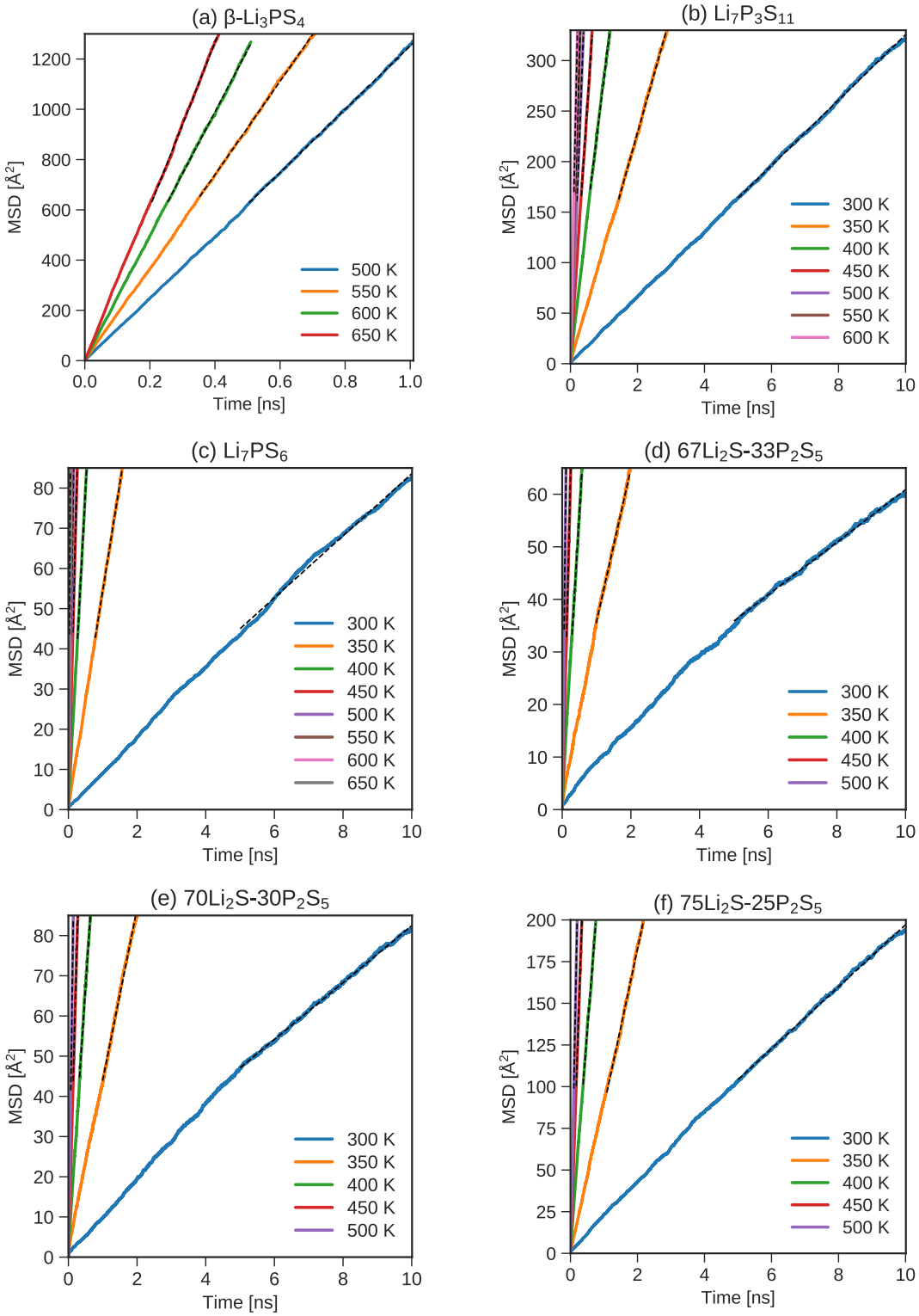

Figure S1: Mean square displacement (MSD) of Li in six compounds with various temperatures are displayed in (a)–(f). Diffusion coefficients are calculated from linear regression drawn with black dashed lines. The fitting time range were half of all simulation run at each temperature.

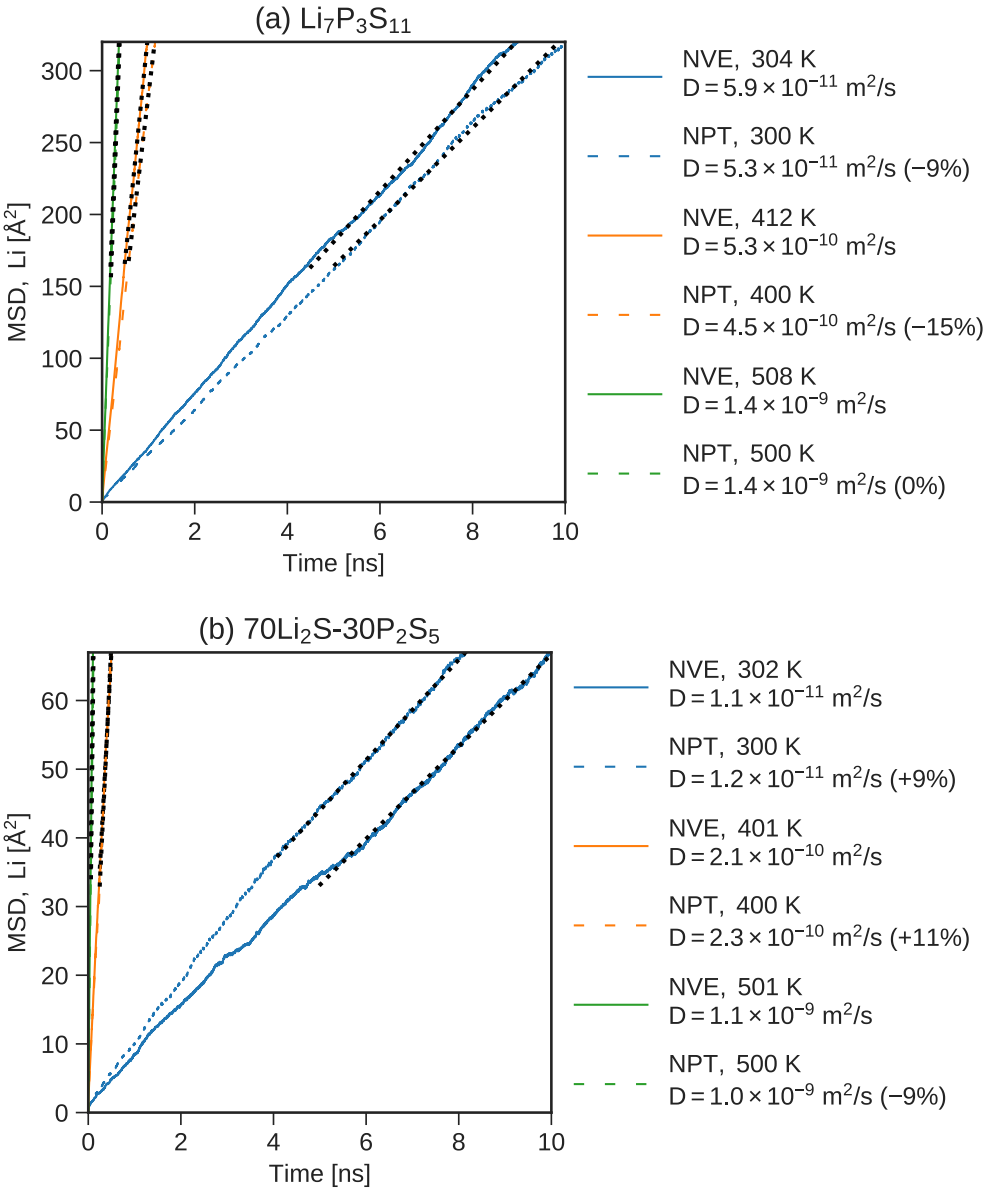

Figure S2: Mean square displacement (MSD) of Li under micro-canonical (NVE) and isobaric-isothermal (NPT) ensembles with various temperatures for (a) $Li_7P_3S_{11}$ crystal and (b) $70Li_2S$-$30P_2S_5$ glass. The temperatures are average values during the simulation run. Diffusion coefficients are calculated from linear regression drawn with black dashed lines. The discrepancy of D between NPT and NVE was shown in parenthesis in %.