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ELECTRONIC SUPPLEMENTARY INFORMATION

Computational design of double-layer cathode coatings in all-solid-state batteries

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Table S1. Interfacial reaction energies and ab-initio molecular dynamics (AIMD) parameters of coating candidates compatible with a $Li_3MnCoNiO_6$ (NCM) cathode.

Materials Project entry id	Composition	Space group	Interfacial reaction energy (meV/atom) with		VASP PAW-PBE potentials
			$Li_3MnCoNiO_6$	$LiMnCoNiO_6$	
mp-1205739	Li_2SnF_6	P-31m	0	0	Li_sv, Sn_d, F
mp-7791	Li_2GeF_6	P4_2/mnm	0	0	Li_sv, Ge_d, F
mp-12829	$LiCaGaF_6$	P-31c	0	0	Li_sv, Ca_sv, Ga_d, F
mp-24199	$LiHF_2$	R-3m	9	0	Li_sv, H, F
mp-1208619	$SrLiGaF_6$	P-31c	0	0	Sr_sv, Li_sv, Ga_d, F
mp-6527	$Na_3Li_3In_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, In_d, F
mp-7603	Li_2TiF_6	P4_2/mnm	0	0	Li_sv, Ti_pv, F
mp-561430	$LiLuF_4$	C2/c	0	0	Li_sv, Lu_3, F
mp-16577	Li_2CaHfF_8	I-4	0	0	Li_sv, Ca_sv, Hf_pv, F
mp-1193269	Li_2SiF_6	P321	0	0	Li_sv, Si, F
mp-29040	$Li_3Zr_4F_{19}$	P-1	0	0	Li_sv, Zr_sv, F
mp-6134	$LiCaAlF_6$	P-31c	0	0	Li_sv, Ca_sv, Al, F
mp-6591	$SrLiAlF_6$	P-31c	0	0	Sr_sv, Li_sv, Al, F
mp-14363	Rb_2LiAsO_4	Cmc2_1	0	0	Rb_sv, Li_sv, As, O
mp-6711	$Na_3Li_3Al_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, Al, F
mp-3700	$LiYF_4$	I4_1/a	0	0	Li_sv, Y_sv, F
mp-4002	Li_2ZrF_6	P-31m	0	0	Li_sv, Zr_sv, F
mp-10103	$LiYbAlF_6$	P-31c	0	0	Li_sv, Yb_2, Al, F
mp-1193222	$LiMgAlF_6$	P321	0	0	Li_sv, Mg_pv, Al, F
mp-15558	Li_3GaF_6	C2/c	0	0	Li_sv, Ga_d, F
mp-18704	$CsLiBeF_4$	P2_1/c	0	0	Cs_sv, Li_sv, Be_sv, F
mp-4622	Li_2BeF_4	R-3	0	0	Li_sv, Be_sv, F
mp-15254	Li_3AlF_6	C2/c	0	0	Li_sv, Al, F
mp-1211087	$LiCdFeF_6$	P-31c	0	0	Li_sv, Cd, Fe_pv, F
mp-14023	$Na_3Li_3Sc_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, Sc_sv, F
mp-543044	$BaLiAlF_6$	P2_1/c	0	0	Ba_sv, Li_sv, Al,

					F
mp-557327	$Na_3Li_3Co_2F_{12}$	Ia-3d	-24.4	0	Na_pv, Li_sv, Co, F
mp-1211093	$LiCaFeF_6$	P-31c	0	0	Li_sv, Ca_sv, Fe_pv, F
mp-8892	$LiInF_4$	Pbcn	0	0	Li_sv, In_d, F
mp-567062	$SrLiFeF_6$	P2_1/c	0	0	Sr_sv, Li_sv, Fe_pv, F
mp-1195868	$LiCeF_5$	I4_1/a	0	0	Li_sv, Ce, F
mp-561280	$Na_3Li_3Fe_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, Fe_pv, F
mp-1196169	$LiThF_5$	I4_1/a	0	0	Li_sv, Th, F
mp-7594	$CsLiF_2$	C2/c	0	0	Cs_sv, Li_sv, F
mp-9308	Li_4ZrF_8	Pnma	0	0	Li_sv, Zr_sv, F
mp-6253	$KLiBeF_4$	P6_3	0	0	K_sv, Li_sv, Be_sv, F
mp-1138	LiF	Fm-3m	0	0	Li_sv, F
mp-776627	Li_3FeF_6	C2/c	0	0	Li_sv, Fe_pv, F
mp-1211435	$KLiTbF_5$	P2_1/c	0	0	K_sv, Li_sv, Tb_3, F
mp-1211585	$KLiGdF_5$	P2_1/c	0	0	K_sv, Li_sv, Gd, F
mp-1211494	$KLiHoF_5$	P2_1/c	0	0	K_sv, Li_sv, Ho_3, F
mp-1211461	$KLiDyF_5$	P2_1/c	0	0	K_sv, Li_sv, Dy_3, F
mp-1211480	$KLiLuF_5$	P2_1/c	0	0	K_sv, Li_sv, Lu_3, F
mp-560518	$RbLi_2Be_2F_7$	P2_1/c	0	0	Rb_sv, Li_sv, Be_sv, F
mp-557798	$K_5Li_2NdF_{10}$	Pnma	0	0	K_sv, Li_sv, Nd_3, F
mp-1196988	$K_5Li_2PrF_{10}$	Pnma	0	0	K_sv, Li_sv, Pr_3, F
mp-1208634	$SrLiCrF_6$	P-31c	0	0	Sr_sv, Li_sv, Cr_pv, F
mp-35759	Li_2NiF_4	Imma	0	0	Li_sv, Ni_pv, F
mp-1212270	$Li_3Lu(NO_3)_6$	P2_1/c	0	0	Li_sv, Lu_3, N, O
mp-561330	$Na_3Li_3Cr_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, Cr_pv, F
mp-28567	$LiBiF_4$	I4_1/a	0	0	Li_sv, Bi, F
mp-1211098	$LiCrCdF_6$	P-31c	0	0	Li_sv, Cr_pv, Cd, F
mp-565544	$BaLiCrF_6$	P2_1/c	0	0	Ba_sv, Li_sv, Cr_pv, F
mp-565468	$LiCaCrF_6$	P-31c	0	0	Li_sv, Ca_sv, Cr_pv, F

mp-720254	$Li_2H_4(SO_4)_3$	Pccn	-23.2	0	Li_sv, H, S, O
mp-29195	$LiPO_3$	P2_1/c	-1	0	Li_sv, P, O
mp-561396	Li_3CrF_6	C2/c	0	0	Li_sv, Cr_pv, F
mp-1211213	$Li_3Tb_3(TeO_6)_2$	Ia-3d	0	0	Li_sv, Tb_3, Te, O
mp-558059	$LiMnF_4$	P2_1/c	-26.8	0	Li_sv, Mn_pv, F
mp-561011	$Li_2Ta_2(OF_2)_3$	P3_121	0	0	Li_sv, Ta_pv, O, F
mp-1211152	$Li_3Eu_3(TeO_6)_2$	Ia-3d	0	0	Li_sv, Eu, Te, O
mp-559129	$Li_3Er(NO_3)_6$	P2_1/c	0	0	Li_sv, Er_3, N, O
mp-1211772	$Li_3Dy(NO_3)_6$	P2_1/c	0	0	Li_sv, Dy_3, N, O
mp-1212059	$Li_3Tm(NO_3)_6$	P2_1/c	0	0	Li_sv, Tm_3, N, O
mp-1212130	$Li_3Tb(NO_3)_6$	P2_1/c	0	0	Li_sv, Tb_3, N, O
mp-1212476	$Li_3Gd(NO_3)_6$	P2_1/c	0	0	Li_sv, Gd, N, O
mp-1212171	$Li_3Y(NO_3)_6$	P2_1/c	0	0	Li_sv, Y_sv, N, O
mp-6726	$CsLiSO_4$	P2_1/c	0	0	Cs_sv, Li_sv, S, O
mp-1212254	$Li_3Yb(NO_3)_6$	P2_1/c	0	0	Li_sv, Yb_2, N, O
mp-540946	$LiReO_4$	P-1	0	0	Li_sv, Re_pv, O
mp-643458	$LiHSO_4$	P2_1/c	-17.6	0	Li_sv, H, S, O
mp-4855	Li_2SeO_4	R-3	0	0	Li_sv, Se, O
mp-30301	$LiClO_4$	Pnma	0	0	Li_sv, Cl, O
mp-14646	$Li_2Mg_2(SO_4)_3$	Pbcn	0	0	Li_sv, Mg_pv, S, O
mp-1194281	$Li_2Co(SO_4)_2$	P2_1/c	0	0	Li_sv, Co, S, O
mp-8180	$LiNO_3$	R-3c	0	0	Li_sv, N, O
mp-4556	Li_2SO_4	P2_1/c	0	0	Li_sv, S, O
mp-6412	$Li_2CuP_2O_7$	C2/c	0	0	Li_sv, Cu_pv, P, O
mp-556229	Li_2MnF_5	C2/c	-20.1	0	Li_sv, Mn_pv, F
mp-767932	Li_2VF_6	P4_2/mnm	6	0	Li_sv, V_pv, F
mp-24610	$LiP(HO_2)_2$	Pna2_1	0	0	Li_sv, P, H, O
mp-558902	$LiMnGaF_6$	P321	0	0	Li_sv, Mn_pv, Ga_d, F
mp-1194965	$Li_2Th(AsO_4)_2$	P2_1/c	0	0	Li_sv, Th, As, O
mp-14484	$KNaLi_2(SO_4)_2$	P2_12_12_1	0	0	K_sv, Na_pv, Li_sv, S, O
mp-6800	$KLiSO_4$	P31c	0	0	K_sv, Li_sv, S, O
mp-25501	$LiCrPO_4F$	P-1	0	0	Li_sv, Cr_pv, P, O, F
mp-9657	$LiAsO_3$	R-3	0	0	Li_sv, As, O
mp-6211	$RbLiSO_4$	P2_1/c	0	0	Rb_sv, Li_sv, S, O
mp-1195620	LiH_2ClO_5	C2/c	0	0	Li_sv, H, Cl, O
mp-560894	$Li_4Be_3P_3ClO_{12}$	P-43n	0	0	Li_sv, Be_sv, P, Cl, O

mp-560072	$Li_4Be_3As_3ClO_{12}$	P-43n	0	0	Li_sv, Be_sv, As, Cl, O
mp-1193172	$Li_2Mn(SO_4)_2$	P2_1/c	0	0	Li_sv, Mn_pv, S, O
mp-555743	$LiZnPO_4$	Cc	0	0	Li_sv, Zn, P, O
mp-1196457	$Li_2B_3O_4F_3$	P2_12_12_1	0	0	Li_sv, B, O, F
mp-25515	$LiFePO_4F$	P-1	0	0	Li_sv, Fe_pv, P, O, F
mp-22983	$LiAlCl_4$	P2_1/c	-21.5	0	Li_sv, Al, Cl
mp-1020705	$Rb_2Li_3B(P_2O_7)_2$	Cmce	0	0	Rb_sv, Li_sv, B, P, O
mp-541190	Cs_2LiVO_4	Cmc2_1	0	0	Cs_sv, Li_sv, V_pv, O
mp-28341	$LiGaCl_4$	P2_1/c	0	0	Li_sv, Ga_d, Cl
mp-18048	$LiZnAsO_4$	R3	0	0	Li_sv, Zn, As, O
mp-554560	$Li_4Be_3P_3BrO_{12}$	P-43n	0	0	Li_sv, Be_sv, P, Br, O
mp-765883	$LiCrPHO_5$	P-1	0	0	Li_sv, Cr_pv, P, H, O
mp-1019778	$K_2Li_3B(P_2O_7)_2$	Cmce	0	0	K_sv, Li_sv, B, P, O
mp-1185263	$LiPaO_3$	Pm-3m	0	0	Li_sv, Pa, O
mp-555001	$LiMnFeF_6$	P321	0	0	Li_sv, Mn_pv, Fe_pv, F
mp-25552	$LiMnPO_4F$	P-1	-10.8	0	Li_sv, Mn_pv, P, O, F
mp-566629	Li_2NiPO_4F	Pnma	0	0	Li_sv, Ni_pv, P, O, F
mp-1222972	$Li_2TiFe(PO_4)_3$	Pna2_1	0	0	Li_sv, Ti_pv, Fe_pv, P, O
mp-565827	$LiMoIO_6$	P2_1	0	0	Li_sv, Mo_pv, I, O
mp-554577	$Li_4P_2O_7$	P2_1/c	0	0	Li_sv, P, O
mp-556065	Li_2MoF_6	P4_2/mnm	0	0	Li_sv, Mo_pv, F
mp-1200209	$Li_2B_6O_9F_2$	Cc	0	0	Li_sv, B, O, F
mp-23626	$RbLi_2(IO_3)_3$	P2_1/c	0	0	Rb_sv, Li_sv, I, O
mp-676109	Li_3InCl_6	C2	0	0	Li_sv, In_d, Cl
mp-1201314	$Li_8Be_6P_7O_{29}$	P31c	-16.1	0	Li_sv, Be_sv, P, O
mp-23384	$LiIO_3$	P4_2/n	0	0	Li_sv, I, O
mp-1020646	$NaLi_2B(PO_4)_2$	P-1	0	0	Na_pv, Li_sv, B, P, O
mp-6113	$LiTiAsO_5$	Pnma	0	0	Li_sv, Ti_pv, As, O
mp-1192681	$LiTcH_6O_7$	P6_3mc	0	0	Li_sv, Tc_pv, H, O
mp-25614	$LiNiPO_4$	Pnma	0	0	Li_sv, Ni_pv, P, O
mp-1222711	Li_2GeTeO_6	R3	0	0	Li_sv, Ge_d, Te, O

mp-1190687	$CsLi_2Cl_3$	Pbcn	0	0	Cs_sv, Li_sv, Cl
mp-615884	$CsLiCrO_4$	P2_1/c	0	0	Cs_sv, Li_sv, Cr_pv, O
mp-771112	$Li_2Mn_3NiO_8$	P4_332	0	0	Li_sv, Mn_pv, Ni_pv, O
mp-676361	Li_3ErCl_6	P321	0	0	Li_sv, Er_3, Cl
mp-1222849	$Li_2ScP_2HO_8$	P2_1	0	0	Li_sv, Sc_sv, P, H, O
mp-9144	$LiAsF_6$	R-3	-17	0	Li_sv, As, F
mp-1080679	$LiTaF_6$	R-3	0	0	Li_sv, Ta_pv, F
mp-3980	$LiSbF_6$	R-3	0	0	Li_sv, Sb, F
mp-12403	$LiBF_4$	P3_121	0	0	Li_sv, B, F
mp-1078799	$LiNbF_6$	R-3	0	0	Li_sv, Nb_pv, F
mp-1185319	LiCl	P6_3mc	0	0	Li_sv, Cl
mp-504360	$Li_9Cr_3P_8O_{29}$	P-3c1	0	0	Li_sv, Cr_pv, P, O
mp-1222810	$Li_2InP_2HO_8$	P2_1	0	0	Li_sv, In_d, P, H, O
mp-24920	Li_2CrO_4	R-3	0	0	Li_sv, Cr_pv, O
mp-1223017	$Li_2TiCr(PO_4)_3$	Pna2_1	0	0	Li_sv, Ti_pv, Cr_pv, P, O
mp-771864	$LiFePHO_5$	P-1	0	0	Li_sv, Fe_pv, P, H, O
mp-1210931	$LiFeCl_4$	P2_1/c	0	0	Li_sv, Fe_pv, Cl
mp-22694	$LiPPbO_4$	Pna2_1	0	0	Li_sv, P, Pb_d, O
mp-1020015	$Li_2B_3PO_8$	P-1	0	0	Li_sv, B, P, O
mp-557177	$Li_2Al(BO_2)_5$	P2_1/c	0	0	Li_sv, Al, B, O
mp-13725	Li_3PO_4	Pmn2_1	0	0	Li_sv, P, O
mp-16828	$Li_3B_7O_{12}$	P-1	0	0	Li_sv, B, O
mp-31788	$Li_3Fe_2(PO_4)_3$	P2_1/c	0	0	Li_sv, Fe_pv, P, O
mp-6565	$Li_3Sc_2(PO_4)_3$	P2_1/c	0	0	Li_sv, Sc_sv, P, O
mp-767473	$Li_2FeP_2HO_8$	P2_1	0	0	Li_sv, Fe_pv, P, H, O
mp-9625	$LiMgPO_4$	Pnma	0	0	Li_sv, Mg_pv, P, O
mp-560104	$LiTaGeO_5$	P2_1/c	0	0	Li_sv, Ta_pv, Ge_d, O
mp-8673	Li_2SnTeO_6	Pnn2	0	0	Li_sv, Sn_d, Te, O
mp-565208	$Li_2U(MoO_5)_2$	P-1	0	0	Li_sv, U, Mo_pv, O
mp-863863	$LiCoPO_4$	Cc	0	0	Li_sv, Co, P, O
mp-8873	$LiGeBO_4$	I-4	0	0	Li_sv, Ge_d, B, O
mp-756117	Li_2TiTeO_6	Pnn2	0	0	Li_sv, Ti_pv, Te, O
mp-585492	$Li_2MgMn_3O_8$	P4_332	0	0	Li_sv, Mg_pv,

					Mn_pv, O
mp-13843	Li_2TeO_4	P4_122	0	0	Li_sv, Te, O
mp-1208625	$SrLiVF_6$	P-31c	0	0	Sr_sv, Li_sv, V_pv, F
mp-6425	$Li_3In_2(PO_4)_3$	R-3	0	0	Li_sv, In_d, P, O
mp-6668	$LiTiPO_5$	Pnma	0	0	Li_sv, Ti_pv, P, O
mp-1222525	$LiAlGeO_4$	R3	0	0	Li_sv, Al, Ge_d, O
mp-772468	$Li_2Cr_3(CoO_6)_2$	Pbcn	-18.2	0	Li_sv, Cr_pv, Co, O
mp-704943	$Li_2Mn_3ZnO_8$	P2_13	0	0	Li_sv, Mn_pv, Zn, O
mp-770932	$LiSbO_3$	C2/m	0	0	Li_sv, Sb, O
mp-761940	$Li_2MnCo_3O_8$	C2	0	0	Li_sv, Mn_pv, Co, O
mp-556256	$Li_3Fe_2(AsO_4)_3$	P2_1/c	0	0	Li_sv, Fe_pv, As, O
mp-9197	Li_3AsO_4	Pmn2_1	0	0	Li_sv, As, O
mp-32316	$Li_2NiGe_3O_8$	P4_332	0	0	Li_sv, Ni_pv, Ge_d, O
mp-25080	Li_2MoO_4	R-3	0	0	Li_sv, Mo_pv, O
mp-23985	LiH_6BrO_7	P6_3mc	0	0	Li_sv, H, Br, O
mp-560297	$LiTaSiO_5$	P2_1/c	0	0	Li_sv, Ta_pv, Si, O
mp-3054	Li_2CO_3	C2/c	0	0	Li_sv, C, O
mp-1222477	$LiVZnO_4$	R3	0	0	Li_sv, V_pv, Zn, O
mp-774082	$Li(CoO_2)_2$	P2_1	0	0	Li_sv, Co, O
mp-19440	$LiVO_3$	C2/c	0	0	Li_sv, V_pv, O
mp-18220	$LiAlSiO_4$	R3	0	0	Li_sv, Al, Si, O
mp-562137	$KLiCO_3$	P2_1/c	0	0	K_sv, Li_sv, C, O
mp-557852	$Na_3Li_3V_2F_{12}$	Ia-3d	0	0	Na_pv, Li_sv, V_pv, F
mp-14364	Cs_2LiAsO_4	Cmc2_1	0	0	Cs_sv, Li_sv, As, O
mp-559533	$NaLiCO_3$	P-6	0	0	Na_pv, Li_sv, C, O
mp-566323	$RbLiMoO_4$	Cc	0	0	Rb_sv, Li_sv, Mo_pv, O
mp-762045	$Li_2Co_3SbO_8$	P4_332	0	0	Li_sv, Co, Sb, O
mp-561689	$CsLiMoO_4$	F-43m	0	0	Cs_sv, Li_sv, Mo_pv, O
mp-18147	$LiGaSiO_4$	R3	0	0	Li_sv, Ga_d, Si, O
mp-1210866	$LiVCdF_6$	P-31c	0	0	Li_sv, V_pv, Cd, F
mp-1211223	$Li_3Yb_3(TeO_6)_2$	Ia-3d	0	0	Li_sv, Yb_2, Te, O

mp-691115	$Li_4Mn_5O_{12}$	C2/c	0	0	Li_sv, Mn_pv, O
mp-31706	$Li_3Cr_2(PO_4)_3$	R-3	0	0	Li_sv, Cr_pv, P, O
mp-18741	$RbLiCrO_4$	P31c	0	0	Rb_sv, Li_sv, Cr_pv, O
mp-1178391	$CsLiWO_4$	I-4	0	0	Cs_sv, Li_sv, W_pv, O
mp-566105	$KLiCrO_4$	Pna2_1	0	0	K_sv, Li_sv, Cr_pv, O
mp-558045	$NaLi_2PO_4$	Pnma	0	0	Na_pv, Li_sv, P, O
mp-9066	$NaLi_2AsO_4$	Pmn2_1	0	0	Na_pv, Li_sv, As, O
mp-1199453	$KLiMoO_4$	P6_3	0	0	K_sv, Li_sv, Mo_pv, O
mp-1176640	$LiMnCrO_4$	Imma	0	0	Li_sv, Mn_pv, Cr_pv, O
mp-1211142	Li_5IO_6	P3_112	0	0	Li_sv, I, O

Table S2. Interfacial reaction energies and AIMD parameters of coating candidates compatible with thiophosphate solid electrolytes.

Materials Project entry id	Composition	Space group	Interfacial reaction energy (meV/atom) with		VASP PAW-PBE potentials
			Li_6PS_5Cl	$Li_{10}GeP_2S_{12}$	
mp-760415	Li_3SbS_4	I-42m	0	0	Li_sv, Sb, S
mp-755664	$Li_2(TaS_2)_3$	P6_322	0	0	Li_sv, Ta_pv, S
mp-1045384	$Li(TiS_2)_2$	Fd-3m	0	0	Li_sv, Ti_pv, S
mp-1192995	$Li_4MnGe_2S_7$	Cc	-6.2	0	Li_sv, Mn_pv, Ge_d, S
mp-1222404	$LiBiS_2$	R-3m	0	0	Li_sv, Bi, S
mp-29410	$Li_2B_2S_5$	Cmcm	0	0	Li_sv, B, S
mp-19755	$Li(TiS_2)_3$	P-3m1	-3.9	0	Li_sv, Ti_pv, S
mp-557962	$SrLiBS_3$	Pnma	-0.4	0	Sr_sv, Li_sv, B, S
mp-1045435	$Li(MnS_2)_2$	Imma	0	0	Li_sv, Mn_pv, S
mp-1222722	$Li(ZrS_2)_2$	P2/m	0	0	Li_sv, Zr_sv, S
mp-853967	LiCuS	Fddd	-3.4	-2.9	Li_sv, Cu_pv, S
mp-1045432	$Li(CrS_2)_2$	Imma	-16.6	-5.8	Li_sv, Cr_pv, S
mp-774459	$Li_8Ti_{16}CuS_{32}$	P1	-1.5	-1.6	Li_sv, Ti_pv, Cu_pv, S
mp-1222482	Li_6AsS_5I	P1	0	0	Li_sv, As, S, I
mp-644419	LiHS	Pmc2_1	0	0	Li_sv, H, S
mp-1188784	Li_2CdSnS_4	Pmn2_1	0	0	Li_sv, Cd, Sn_d, S
mp-753720	Li_3BiS_3	R-3	0	0	Li_sv, Bi, S
mp-3647	$LiGaS_2$	Pna2_1	0	0	Li_sv, Ga_d, S
mp-573030	Cs_2LiNbS_4	P-1	0	0	Cs_sv, Li_sv, Nb_pv, S
mp-1222735	Li_2VCrS_4	C2/m	0	0	Li_sv, V_pv, Cr_pv, S
mp-767171	$Li_5(NbS_2)_7$	C2/c	0	0	Li_sv, Nb_pv, S
mp-755309	Li_3NbS_4	P-43m	-0.1	-0.1	Li_sv, Nb_pv, S
mp-754856	$Li_7Y_7ZrS_{16}$	P2/m	0	0	Li_sv, Y_sv, Zr_sv, S
mp-1225894	$CsLiMnS_2$	I-4m2	-14	-3.5	Cs_sv, Li_sv, Mn_pv, S
mp-28471	Li_3AsS_3	Pna2_1	0	0	Li_sv, As, S
mp-559814	$Li_5SbS_3I_2$	Pnnm	0	0	Li_sv, Sb, S, I
mp-1190364	Li_2SnS_3	C2/c	-8.4	0	Li_sv, Sn_d, S
mp-767218	$Li_9(NbS_2)_{14}$	P-1	-0.9	0	Li_sv, Nb_pv, S
mp-557892	$BaLi(BS_2)_3$	Cc	-1.2	0	Ba_sv, Li_sv, B, S
mp-644271	LiHS	Ama2	0	0	Li_sv, H, S
mp-1105291	Li_2MnSnS_4	Pc	0	0	Li_sv, Mn_pv, Sn_d, S
mp-766506	Li_3CuS_2	Ia-3	0	0	Li_sv, Cu_pv, S
mp-1211176	Li_6AsS_5I	Cc	0	0	Li_sv, As, S, I
mp-1079885	$LiSbS_2$	C2/c	-0.2	0	Li_sv, Sb, S

mp-753946	$LiCoS_2$	P-3m1	0	0	Li_sv, Co, S
mp-554076	$BaLiBS_3$	P2_1/c	-1.9	-2	Ba_sv, Li_sv, B, S
mp-767165	K_2LiVS_4	Cc	-2.2	0	K_sv, Li_sv, V_pv, S
mp-1210804	Li_2ZnGeS_4	Pmn2_1	-3.9	0	Li_sv, Zn, Ge_d, S
mp-1222582	Li_4GeS_4	Pna2_1	0	0	Li_sv, Ge_d, S
mp-1191903	$Li_2In_2SiS_6$	Cc	-12.4	-0.9	Li_sv, In_d, Si, S
mp-1206881	$LiTaS_2$	P6_3/mmc	-13.6	-13.7	Li_sv, Ta_pv, S
mp-558815	Rb_2LiVS_4	Fddd	0	0	Rb_sv, Li_sv, V_pv, S
mp-1021497	Li_2SiSnS_4	I-42m	0	0	Li_sv, Si, Sn_d, S
mp-1195603	Li_2MnSnS_4	Pna2_1	0	0	Li_sv, Mn_pv, Sn_d, S
mp-5614	Li_3BS_3	Pnma	0	0	Li_sv, B, S
mp-1194339	Li_3SbS_3	Pna2_1	0	0	Li_sv, Sb, S
mp-756490	Li_6MnS_4	P4_2/nmc	-9.2	-9.2	Li_sv, Mn_pv, S
mp-1176780	$LiCoS_2$	P2/c	0	0	Li_sv, Co, S
mp-14591	$LiSbS_2$	R-3	-0.2	0	Li_sv, Sb, S
mp-554395	$Na_5Li_3(TiS_4)_2$	C2/c	-5.5	0	Na_pv, Li_sv, Ti_pv, S
mp-33526	$LiBiS_2$	I4_1/amd	0	0	Li_sv, Bi, S
mp-19896	Li_2GePbS_4	I-42m	-5.6	0	Li_sv, Ge_d, Pb_d, S
mp-558219	$SrLi(BS_2)_3$	Cc	-2.9	-0.4	Sr_sv, Li_sv, B, S
mp-1188392	$LiInS_2$	Pna2_1	0	0	Li_sv, In_d, S
mp-1189383	Li_2CdGeS_4	Pmn2_1	0	0	Li_sv, Cd, Ge_d, S
mp-532413	$Li_5B_7S_{13}$	Cc	-1.2	0	Li_sv, B, S
mp-510338	Cs_2LiVS_4	Fddd	0	0	Cs_sv, Li_sv, V_pv, S
mp-7543	$LiVS_2$	P-3m1	0	0	Li_sv, V_pv, S
mp-556085	Rb_2LiTaS_4	P-1	0	0	Rb_sv, Li_sv, Ta_pv, S
mp-553962	Rb_2LiNbS_4	P-1	0	0	Rb_sv, Li_sv, Nb_pv, S
mp-29829	LiAuS	Fddd	-3.8	0	Li_sv, Au, S
mp-555186	Li_2ZnSnS_4	Pc	0	0	Li_sv, Zn, Sn_d, S
mp-558731	Li_2TeS_3	P2_1/c	0	0	Li_sv, Te, S
mp-7936	$LiNbS_2$	P6_3/mmc	-0.2	-0.2	Li_sv, Nb_pv, S
mp-1001786	$LiScS_2$	R-3m	0	0	Li_sv, Sc_sv, S
mp-1001784	$LiTiS_2$	R-3m	-5.8	-5.9	Li_sv, Ti_pv, S
mp-756006	LiSbS	P1	0	0	Li_sv, Sb, S
mp-559238	$CsLi_2BS_3$	Pnma	0	0	Cs_sv, Li_sv, B, S
mp-1211182	$LiCr_4InS_8$	F-43m	-18.3	-4.6	Li_sv, Cr_pv, In_d, S
mp-767137	$Li_3Sb_{17}S_{27}$	P1	-5.5	0	Li_sv, Sb, S
mp-15999	Li_3AuS_2	Ibam	0	0	Li_sv, Au, S
mp-1153	Li_2S	Fm-3m	0	0	Li_sv, S

mp-4226	$LiCrS_2$	P-3m1	0	0	Li_sv, Cr_pv, S
mp-766540	Li_4TiS_4	Pnma	-0.6	0	Li_sv, Ti_pv, S
mp-30959	LiHS	P4_2/mbc	0	0	Li_sv, H, S
mp-769032	Li_3NbS_4	I-43m	-0.1	-0.1	Li_sv, Nb_pv, S
mp-1106183	$LiAlS_2$	Pna2_1	0	0	Li_sv, Al, S
mp-756316	Li_3SbS_4	Pmn2_1	0	0	Li_sv, Sb, S
mp-1191476	$Li_2In_2GeS_6$	Cc	-15	0	Li_sv, In_d, Ge_d, S
mp-767088	$Li_3Sb_{11}S_{18}$	P1	-5.2	0	Li_sv, Sb, S
mp-555874	$LiAsS_2$	Cc	-3.4	0	Li_sv, As, S
mp-861182	Li_4TiS_4	I-42m	-0.6	0	Li_sv, Ti_pv, S
mp-995393	LiS_4	P1	0	0	Li_sv, S
mp-760375	Li_3VS_4	P-43m	0	0	Li_sv, V_pv, S
mp-753546	Li_8TiS_6	P6_3cm	0	0	Li_sv, Ti_pv, S
mp-1177695	Li_3CuS_2	Ibam	0	0	Li_sv, Cu_pv, S
mp-1225887	$CsLiZnS_2$	I-4m2	-12.3	-1.5	Cs_sv, Li_sv, Zn, S
mp-756198	$Li_{14}Mn_2S_9$	P-3	-8.3	-8.3	Li_sv, Mn_pv, S
mp-1220728	$NaLi(AsS_2)_2$	P1	-3	0	Na_pv, Li_sv, As, S
mp-756811	Li_4TiS_4	Cmcm	-0.6	0	Li_sv, Ti_pv, S
mp-33363	$LiYS_2$	I4_1/amd	0	0	Li_sv, Y_sv, S
mp-767467	$Li_7Y_7Zr_9S_{32}$	P1	0	0	Li_sv, Y_sv, Zr_sv, S
mp-15788	$LiYS_2$	R-3m	0	0	Li_sv, Y_sv, S

Table S3. Oxidation voltage limit of coatings compatible with NCM cathode.

MP entry id	Composition	Oxidation voltage (eV)
mp-556256	$Li_3Fe_2(AsO_4)_3$	4.11
mp-676109	Li_3InCl_6	4.42
mp-676361	Li_3ErCl_6	4.27
mp-1210931	$LiFeCl_4$	4.14
mp-561396	Li_3CrF_6	5.01
mp-1195868	$LiCeF_5$	6.43
mp-28341	$LiGaCl_4$	4.41
mp-9308	Li_4ZrF_8	6.38
mp-31788	$Li_3Fe_2(PO_4)_3$	4.18
mp-12403	$LiBF_4$	7.10
mp-6425	$Li_3In_2(PO_4)_3$	4.17

Table S4. Training and validation errors on energies and components of force for the moment tensor potentials after completion of LOTF-MD on 8 benchmark materials. The training errors were computed for all configurations used to train the potentials. For each structure, the validation set contains 100 snapshots (100 ps between each snapshot) from a 10 ns LOTF-MD trajectory for each structure at 700 K.

Type	MP entry id	Composition	Fitting Mean absolute error		Validation Mean absolute error	
			energy MAE meV/atom	force MAE meV/Å	energy MAE meV/atom	force MAE meV/Å
Sulfide-based electrolyte coating	mp-1045384	$Li(TiS_2)_2$	2.43	137.21	4.23	53.33
	mp-14591	$LiSbS_2$	2.97	129.16	7.44	69.43
	mp-532413	$Li_5B_7S_{13}$	1.5	124.65	5.47	50.63
	mp-753720	Li_3BiS_3	1.92	83.38	3.4	44.24
NCM-cathode coating	mp-1195868	$LiCeF_5$	1.25	48.05	19.07	58.46
	mp-1210931	$LiFeCl_4$	5.55	85.3	5.64	76.17
	mp-561396	Li_3CrF_6	0.76	34.52	4.15	45.13
	mp-6425	$Li_3In_2(PO_4)_3$	1.16	59.62	7.45	52.88
Average			2.19	87.74	7.11	56.28

Section S1: Estimation of room temperature ionic conductivity cutoff for coating materials screening

Using the Einstein-Nernst equation, we relate the diffusion activation energy to ionic conductivity as a function of temperature:

$$\sigma(T) = \frac{ne^2z^2}{k_BT}D(T) = \frac{ne^2z^2}{k_BT}(fa^2\nu_0)e^{-\frac{E_a}{k_BT}} \quad \text{(S1)}$$

where n is the volume density of the diffusing species, e is the unit electron charge, z is the charge of the ionic conductor (here 1 for Li^+), k_B is Boltzmann's constant, f is the diffusion correlation coefficient, a is the average hopping distance, ν_0 is the Li attempt frequency. We take f = 1, a = 3 Å, $\nu_0 = 10^{12}\ s^{-1}$ to be typical for Li ion diffusion and n = 0.012 $Å^{-3}$ (approximated by the average Li concentration across all simulated structures). Using an activation energy cutoff of 0.7 eV, we calculate the lower bound of ionic conductivity at 300K to be 6.12×10^{-5} mS/cm, which is close to the lower bound of acceptable conductivities for thin coating layers we calculated in our previous work.[1]

Reference:

1. C. Wang, K. Aoyagi, P. Wisesa and T. Mueller, *Chem Mater*, 2020, **32**, 3741-3752.