Supplementary Material

A Paradigm for Systematic Screening and Evaluation of Artificial Solid-Electrolyte Interfaces for Lithium Metal Anodes: A Computational Study of Binary Selenides

Wenshan Xiao[a], Mingwei Wu[a], Huan Wang[a], Qiu He[a,c *], Yan Zhao[a,b,c *]

[a] *International School of Materials Science and Engineering, State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Hubei 430070, China.*

[b] *The Institute of Technological Sciences, Wuhan University, Hubei, Wuhan 430072, China.*

[c] *College of Materials Science and Engineering, Sichuan University, Chengdu 610065, China*

[*] Corresponding author. E-mail address: hq5220@whut.edu.cn (Q. H.)

yan2000@whut.edu.cn (Y. Z.)

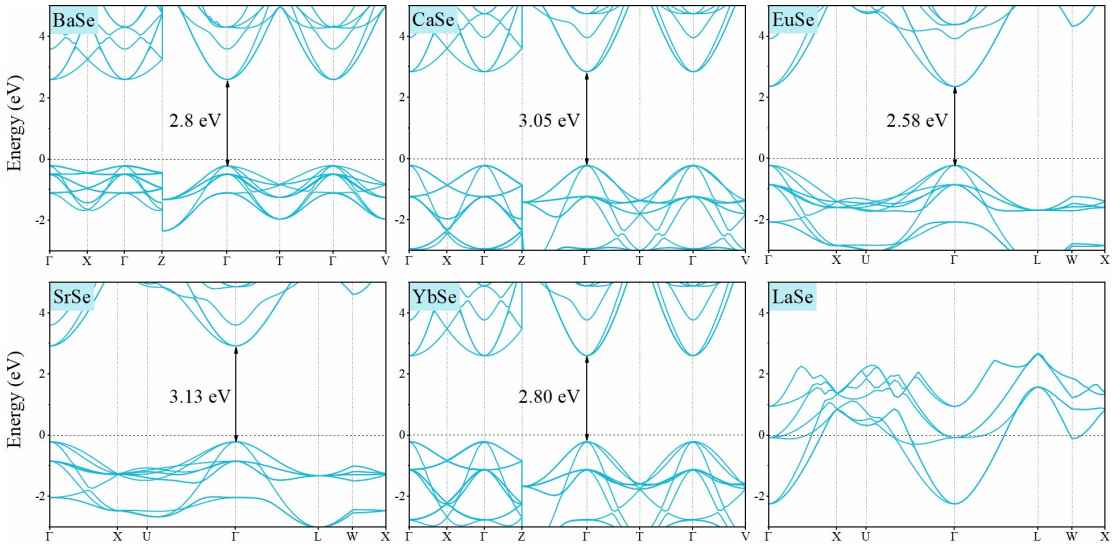


Fig. S1. Band structures of six lithium-stable selenides.

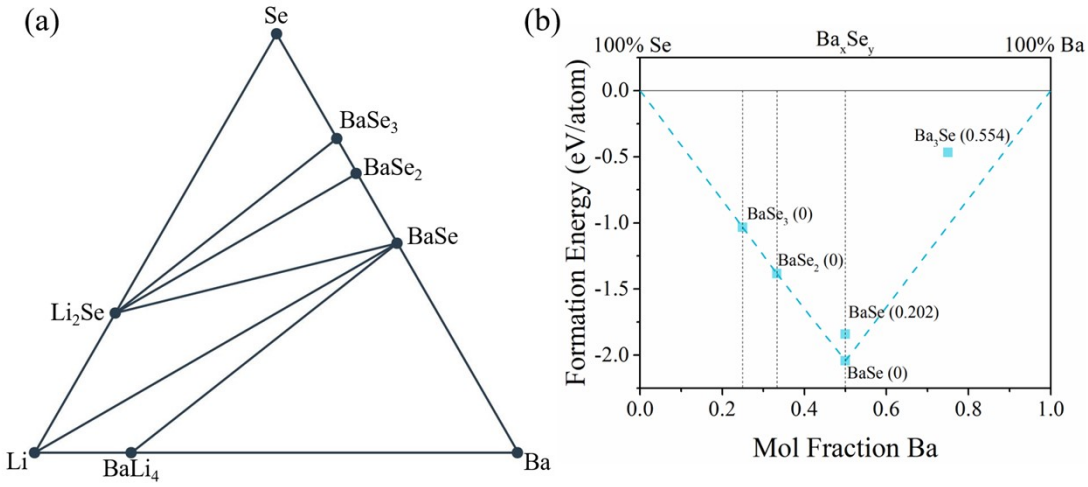


Fig. S2. (a) Phase diagram of Li-Ba-Se. (b) Convex hull diagram of Ba_xSe_y with energies of five Ba-Se phases above the convex hull marked.

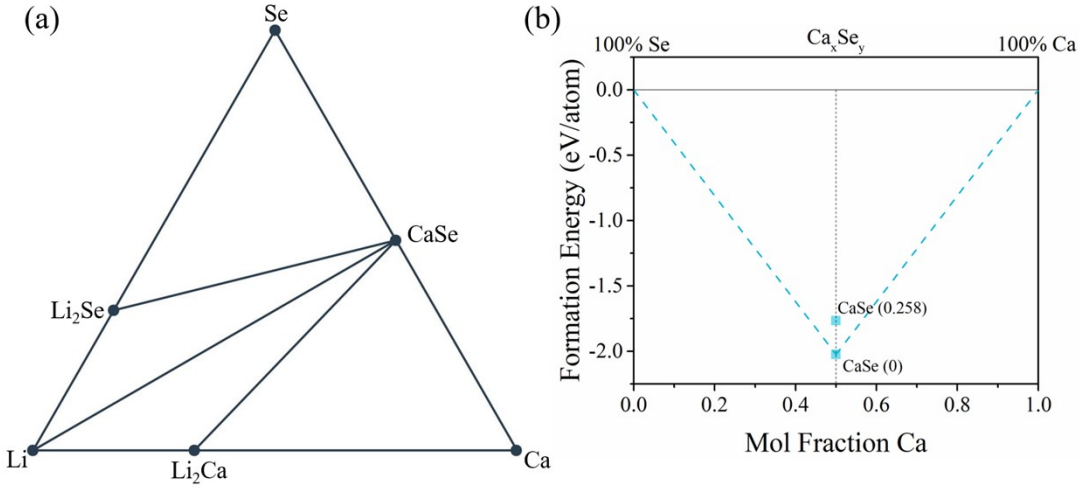


Fig. S3. (a) Phase diagram of Li-Ca-Se. (b) Convex hull diagram of Ca_xSe_y with energies of two Ca-Se phases above the convex hull marked.

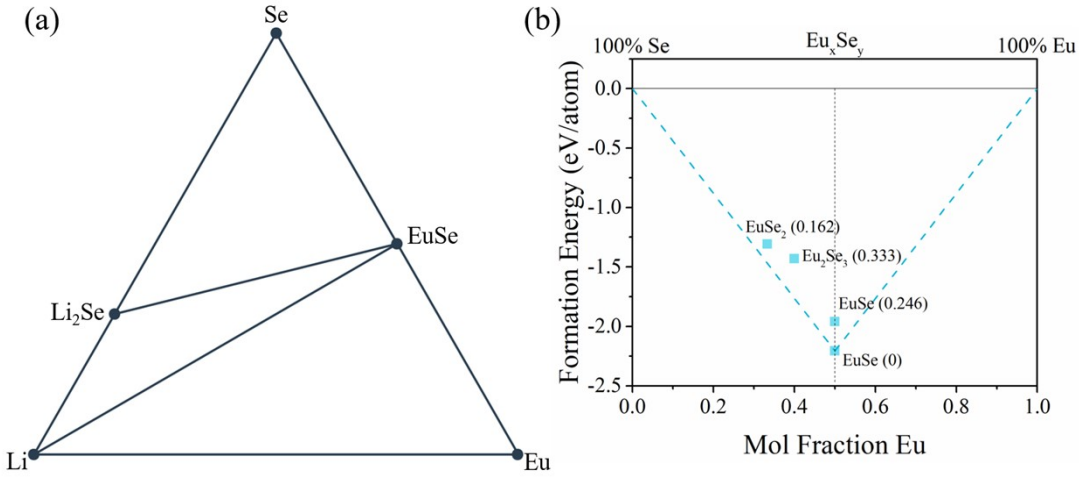


Fig. S4. (a) Phase diagram of Li-Eu-Se. (b) Convex hull diagram of Eu_xSe_y with energies of four Eu-Se phases above the convex hull marked.

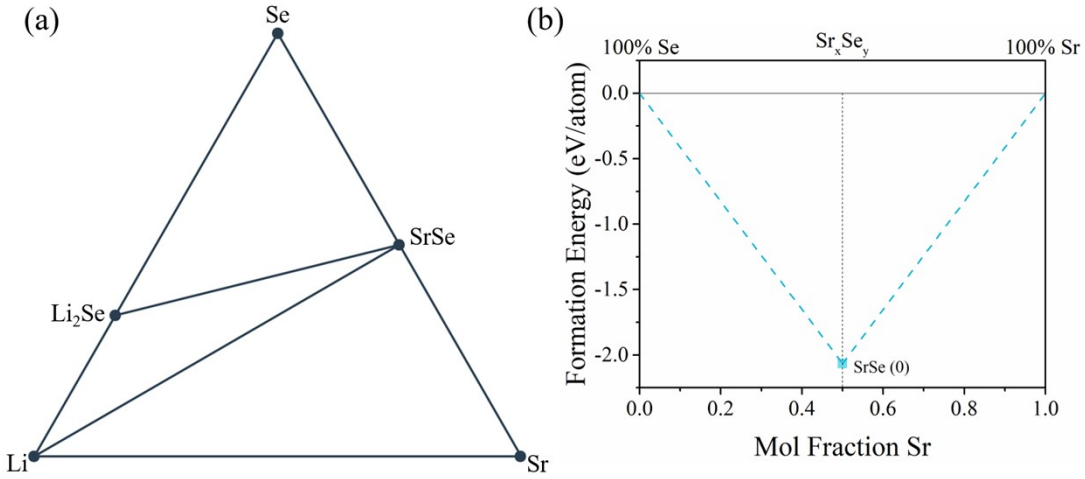


Fig. S5. (a) Phase diagram of Li-Sr-Se. (b) Convex hull diagram of Sr_xSe_y with the energy of SrSe above the convex hull marked.

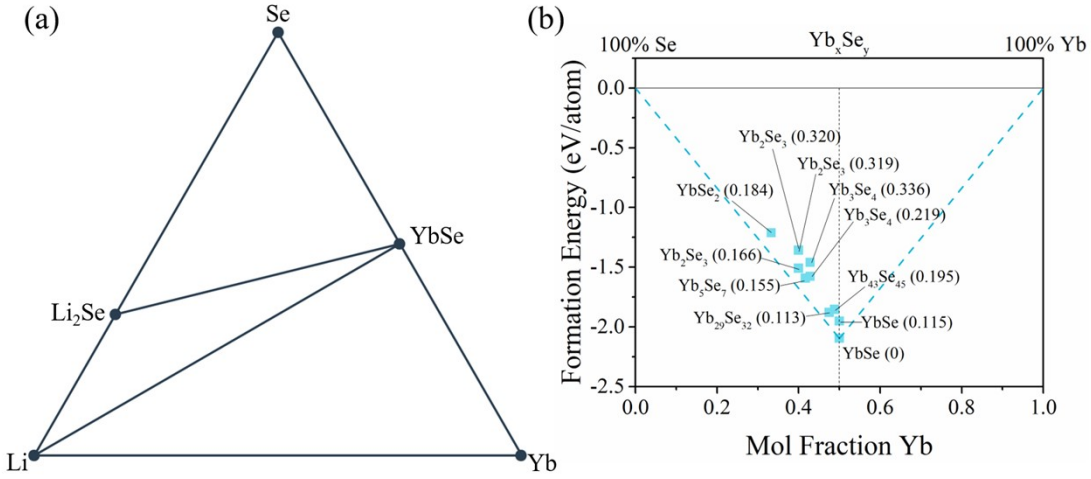


Fig. S6. (a) Phase diagram of Li-Yb-Se. (b) Convex hull diagram of Yb_xSe_y with energies of eleven Yb-Se phases above the convex hull marked.

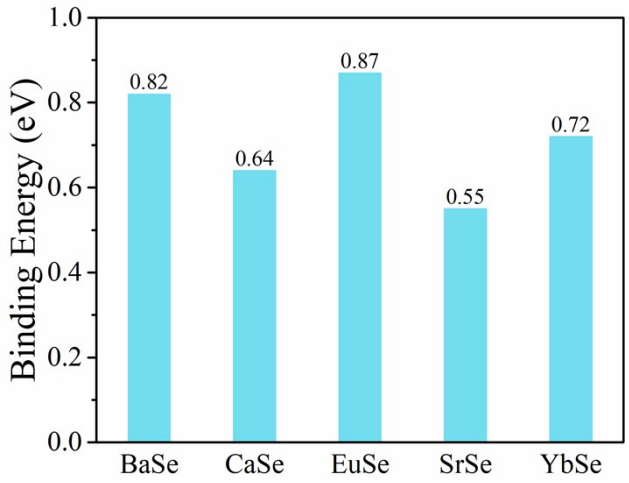


Fig. S7. Binding energies between the Li atom and selenides at site 2.

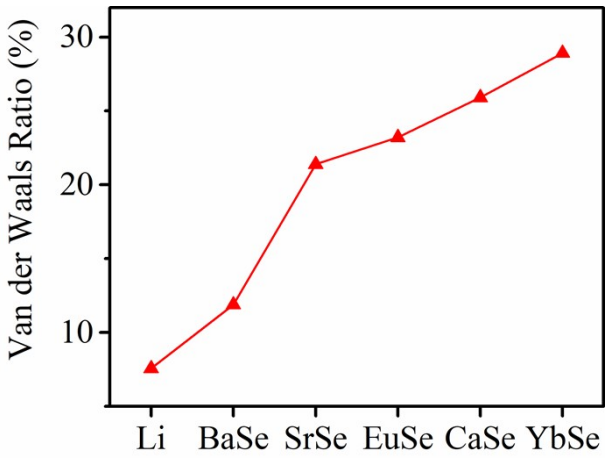
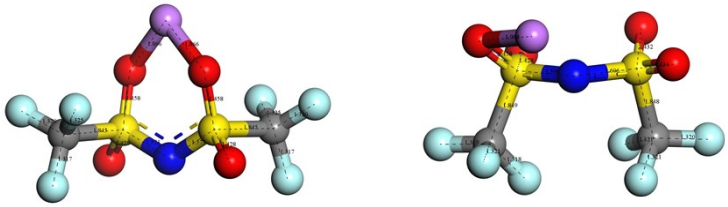


Fig. S8. Ratios of vdW interaction of Li ion with lithium metal/selenides.



0 8.88

Fig. S9. Isomers of LiTFSI. The relative energy (kcal/mol) is shown below each isomer.

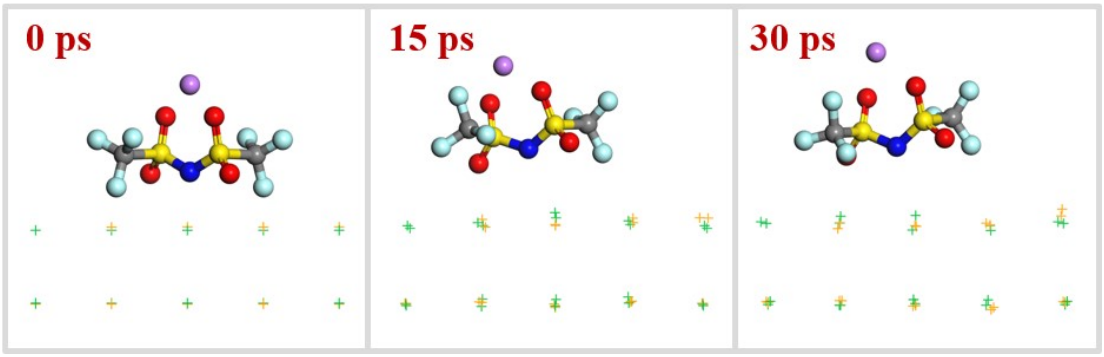


Fig. S10. Sequence of LiTFSI behavior on YbSe surface in DOL/DME mixture obtained from AIMD simulation (LiTFSI is not decomposed).

Table S1. The cathodic limits, lithiation products, and reaction energy E_D of binary selenides.

Compound	Phase equilibria at cathodic limit	Cathodic limit ref. to Li/Li^+ (V)	Phase equilibria with Li metal	E_D per Li (eV)
BeSe	Be, Li_2Se	1.098	Be, Li_2Se	-1.098
BSe_2	B, $Li_2B_2Se_5$	1.800	LiB, $Li_2B_2Se_5$	-1.509
CSe_2	C, Li_2Se	2.395	$LiC1_2$, Li_2Se	-2.347
Na_2Se	Na, NaLiSe	0.315	Na, Li_2Se	-0.288
MgSe	$LiMg_{149}$, Li_2Se	0.649	Li_5Mg, Li_2Se	-0.201
Al_2Se_3	Al, $LiAlSe_2$	1.557	Li_2Al, Li_2Se	-0.812
$SiSe_2$	Si, Li_2Se	1.553	$Li_{21}Si_5$, Li_2Se	-0.893
P_2Se_5	PSe, Li_7PSe_6	1.954	Li_3P, Li_2Se	-1.526
K_2Se	K, KLiSe	0.350	K, Li_2Se	-0.246
CaSe	Li_2Ca, Li_2Se	-0.057	Li_2Ca, Li_2Se	0.057
Sc_2Se_3	ScSe, Li_2Se	0.803	Sc, Li_2Se	-0.469
$TiSe_2$	$Li(TiSe_2)_3$	1.575	Ti, Li_2Se	-1.057
V_2Se_9	VSe_2, Li_2Se	1.855	V, Li_2Se	-1.640
$CrSe_2$	Cr_5Se_8, Li_2Se	1.885	Cr, Li_2Se	-1.550
$MnSe_2$	$LiMn_9Se_{10}$, Li_2Se	1.821	Mn, Li_2Se	-0.952
$FeSe_2$	FeSe, Li_2Se	1.630	Fe, Li_2Se	-1.625
$CoSe_2$	Co_3Se_4, Li_2Se	1.726	Co, Li_2Se	-1.668
$NiSe_2$	Ni_3Se_4, Li_2Se	1.888	Ni, Li_2Se	-1.711
CuSe	Cu_3Se_2, Li_2Se	1.854	$LiCu_3$, Li_2Se	-1.535
ZnSe	Zn, Li_2Se	1.175	LiZn, Li_2Se	-0.929
Ga_2Se_3	GaSe, $LiGaSe_2$	1.900	Li_2Ga, Li_2Se	-1.021
$GeSe_2$	GeSe, Li_2Se	1.720	$Li_{15}Ge_4$, Li_2Se	-1.077
As_2Se_3	As, $LiAsSe_2$	1.821	Li_3As, Li_2Se	-1.371
RbSe	RbLiSe, Li_2Se	1.684	Rb, Li_2Se	-0.963
SrSe	Sr, Li_2Se	-0.172	Sr, Li_2Se	0.172
Y_2Se_3	YSe, $LiYSe_2$	0.845	Y, Li_2Se	-0.311
$ZrSe_2$	$LiZrSe_2$, Li_2Se	1.440	Zr, Li_2Se	-0.894
Nb_2Se_9	$NbSe_2$, Li_2Se	1.833	Nb, Li_2Se	-1.578
$MoSe_2$	$Li_7(Mo_3Se_4)_4$, Li_2Se	1.389	Mo, Li_2Se	-1.386
$RuSe_2$	Ru, Li_2Se	1.572	Ru, Li_2Se	-1.572
$RhSe_2$	Rh_2Se_3, Li_2Se	1.623	LiRh, Li_2Se	-1.331
$PdSe_2$	PdSe, Li_2Se	1.793	$Li_{15}Pd_4$, Li_2Se	-1.032
Ag_2Se	Ag, Li_2Se	1.790	Li_3Ag, Li_2Se	-0.605
$CdSe_2$	CdSe, Li_2Se	2.011	Li_3Cd, Li_2Se	-1.041
In_2Se_3	InSe, $LiInSe_2$	1.909	$Li_{13}In_3$, Li_2Se	-0.747
$SnSe_2$	SnSe, Li_2SnSe_3	1.907	$Li_{17}Sn_4$, Li_2Se	-1.019
Sb_2Se_3	Sb, $LiSbSe_2$	1.696	Li_3Sb, Li_2Se	-1.296
$TeSe_2$	$LiTe_3$, Li_2Se	1.956	Li_2Te, Li_2Se	-1.847
Cs_2Se	Cs, Li_2Se	0.319	Cs, Li_2Se	-0.319
BaSe	$BaLi_4$, Li_2Se	-0.052	$BaLi_4$, Li_2Se	0.052

La_2Se_3	LaSe, Li_2Se	0.804	LaSe, Li_2Se	-0.804
LaSe	La, Li_2Se	-0.104	La, Li_2Se	0.104
$CeSe_2$	$Ce_{10}Se_{19}$, Li_2Se	1.886	Ce, Li_2Se	-0.689
Pr_2Se_3	$Pr_{27}Se_{40}$, Li_2Se	1.223	Pr, Li_2Se	-0.319
Nd_2Se_3	NdSe, Li_2Se	0.848	Nd, Li_2Se	-0.323
Pm_2Se_3	PmSe, Li_2Se	0.805	Pm, Li_2Se	-0.311
Sm_2Se_3	SmSe, $LiSmSe_2$	0.811	Sm, Li_2Se	-0.317
$EuSe_2$	EuSe, Li_2Se	1.896	EuSe, Li_2Se	-1.896
EuSe	Eu, Li_2Se	-0.135	Eu, Li_2Se	0.135
Gd_2Se_3	GdSe, $LiGdSe_2$	1.032	Gd, Li_2Se	-0.313
Tb_2Se_3	TbSe, $LiTbSe_2$	0.937	Tb, Li_2Se	-0.331
Dy_2Se_3	DySe, $LiDySe_2$	0.962	Dy, Li_2Se	-0.341
Ho_2Se_3	HoSe, $LiHoSe_2$	0.811	Ho, Li_2Se	-0.325
Er_2Se_3	ErSe, $LiErSe_2$	0.772	Er, Li_2Se	-0.329
Tm_2Se_3	TmSe, Li_2Se	0.528	Tm, Li_2Se	-0.335
$YbSe_2$	YbSe, Li_2Se	2.167	YbSe, Li_2Se	-2.167
YbSe	Yb, Li_2Se	-0.195	Yb, Li_2Se	0.195
Lu_2Se_3	LuSe, Li_2Se	0.438	Lu, Li_2Se	-0.350
$HfSe_2$	$LiHfSe_2$, Li_2Se	1.150	Hf, Li_2Se	-0.882
$TaSe_3$	$TaSe_2$, Li_2Se	1.812	Ta, Li_2Se	-1.455
WSe_2	W, Li_2Se	1.473	W, Li_2Se	-1.473
$ReSe_2$	Re, Li_2Se	1.591	Re, Li_2Se	-1.591
$OsSe_2$	Os, Li_2Se	1.758	Os, Li_2Se	-1.758
$IrSe_2$	Ir, Li_2Se	1.634	LiIr, Li_2Se	-1.414
$PtSe_2$	Pt_5Se_4, Li_2Se	1.633	Li_3Pt, Li_2Se	-1.153
AuSe	Au, Li_2Se	1.790	$Li_{15}Au_4$, Li_2Se	-0.914
HgSe	Hg, Li_2Se	1.685	Li_3Hg, Li_2Se	-0.898
Tl_2Se_3	TlSe, Li_2Se	1.921	Li_3Tl, Li_2Se	-1.012
$PbSe_2$	PbSe, Li_2Se	1.972	$Li_{17}Pb_4$, Li_2Se	-0.985
$BiSe_2$	Bi_2Se_3, Li_2Se	2.014	Li_3Bi, Li_2Se	-1.263
$AcSe_3$	Ac, Li_2Se	1.193	Ac, Li_2Se	-1.193
$ThSe_2$	Th_7Se_{12}, Li_2Se	1.092	Th, Li_2Se	-0.593
$PaSe_3$	Pa, Li_2Se	1.322	Pa, Li_2Se	-1.322
USe_3	USe_2, Li_2Se	1.732	U, Li_2Se	-1.269
Np_2Se_5	Np_3Se_5, Li_2Se	1.732	Np, Li_2Se	-1.209
$PuSe_2$	Pu_2Se_3, Li_2Se	1.828	Pu, Li_2Se	-0.914
$Li_2B_2Se_5$	B, Li_2Se	1.788	LiB, Li_2Se	-1.500
Li_8SeN_2	Li_3N, Li_2Se	-0.009	Li_3N, Li_2Se	0.009
NaLiSe	Na, Li_2Se	0.281	Na, Li_2Se	-0.281
$LiAlSe_2$	Al, Li_2Se	1.069	Li_2Al, Li_2Se	-0.739
Li_7PSe_6	P, Li_2Se	1.851	Li_3P, Li_2Se	-1.505
KLiSe	K, Li_2Se	0.141	K, Li_2Se	-0.141
$Li(TiSe_2)_3$	$LiTiSe_2$, Li_2Se	1.557	Ti, Li_2Se	-1.010
$LiTiSe_2$	Ti_3Se_4, Li_2Se	1.365	Ti, Li_2Se	-1.365

$LiMn_9Se_{10}$	Mn, Li_2Se	1.528	Mn, Li_2Se	-1.528
$LiGaSe_2$	GaSe, Li_2Se	1.322	Li_2Ga, Li_2Se	-0.964
$LiAsSe_2$	As, Li_2Se	1.783	Li_3As, Li_2Se	-1.366
RbLiSe	Rb, Li_2Se	0.241	Rb, Li_2Se	-0.241
$LiYSe_2$	YSe, Li_2Se	0.498	Y, Li_2Se	-0.253
$LiZrSe_2$	ZrSe, Li_2Se	1.048	Zr, Li_2Se	-0.712
$LiNbSe_2$	Nb, Li_2Se	1.089	Nb, Li_2Se	-1.089
$Li_7(Mo_3Se_4)_4$	Mo, Li_2Se	1.383	Mo, Li_2Se	-1.383
$LiInSe_2$	In, Li_2Se	1.360	$Li_{13}In_3$, Li_2Se	-0.706
Li_2SnSe_3	SnSe, Li_2Se	1.723	$Li_{17}Sn_4$, Li_2Se	-0.997
$LiSbSe_2$	Sb, Li_2Se	1.672	Li_3Sb, Li_2Se	-1.293
$LiSmSe_2$	SmSe, Li_2Se	0.767	Sm, Li_2Se	-0.310
$LiTbSe_2$	TbSe, Li_2Se	0.537	Tb, Li_2Se	-0.264
$LiDySe_2$	DySe, Li_2Se	0.492	Dy, Li_2Se	-0.263
$LiHoSe_2$	HoSe, Li_2Se	0.439	Ho, Li_2Se	-0.263
$LiErSe_2$	ErSe, Li_2Se	0.385	Er, Li_2Se	-0.264
$LiHfSe_2$	Hf_2Se, Li_2Se	0.849	Hf, Li_2Se	-0.792

Table S2. Bond angle and bond length changes near the adsorption site of five selenides before and after adsorption of Li^+.

Species		Length 1	Length 2	Length 3	Length 4	Δ Length	Angle	Δ Angle
BaSe	Before	3.343	3.343	3.344	3.344	0.074	90.018	0.234
	After	3.43	3.418	3.405	3.416		89.784	
CaSe	Before	2.985	2.985	2.985	2.985	0.061	89.889	0.041
	After	3.045	3.047	3.046	3.044		89.848	
EuSe	Before	3.06	3.06	3.06	3.06	0.067	89.952	0.089
	After	3.127	3.124	3.126	3.129		89.863	
SrSe	Before	3.152	3.152	3.152	3.152	0.060	89.95	0.047
	After	3.212	3.207	3.212	3.216		89.997	
YbSe	Before	2.956	2.956	2.956	2.956	0.058	89.886	0.025
	After	3.016	3.011	3.011	3.017		89.861	