Supporting Information

High-capacity cathodes for magnesium lithium chlorine tri-ion batteries through chloride intercalation in layered MoS_2: a computational study

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Table S1. Voltages (*v.s.* Li) and capacities in the Li_xMoS_2 system

Compositions	Voltage *v.s.* Li (V)	Capacity ($mAhg^{-1}$)
$Li_{0.25}MoS_2$	2.5	41.4
$Li_{0.5}MoS_2$	1.54	82.0
$Li_{0.75}MoS_2$	1.49	121.6
$LiMoS_2$	1.35	160.5
$Li_{1.5}MoS_2$	0.37	-

Table S2. Voltages (*v.s.* Mg) and capacities in the Mg_xMoS_2 or hybrid Mg^{2+}/Li^+ compounds with stable *dT* structures.

Compositions	Voltage *v.s.* Mg (V)	Capacity ($mAhg^{-1}$)
$Mg_{0.25}MoS_2$	0.65	80.7
$Mg_{0.5}MoS_2$	0.18	155.8
$Mg_{0.75}MoS_2$	0.06	-
$Li_{0.25}Mg_{0.25}MoS_2$	1.08	119.8
$Li_{0.5}Mg_{0.25}MoS_2$	0.9	160.0

Table S3. Voltage (*v.s.* Mg) and capacities in $Li_xMg_yMoS_2Cl_{0.5}$ compounds with stable *dT* structures.

Compositions	Voltage *v.s.* Mg (V)	Capacity ($mAhg^{-1}$)
$Li_{0.5}MoS_2Cl_{0.5}$	2.4	147.9
$Mg_{0.25}MoS_2Cl_{0.5}$	1.9	145.9
$Li_{0.25}Mg_{0.25}MoS_2Cl_{0.5}$	1.1	180.6
$Li_{0.5}Mg_{0.25}MoS_2Cl_{0.5}$	0.92	214.7
$LiMoS_2Cl_{0.5}$	0.85	217.6
$Mg_{0.5}MoS_2Cl_{0.5}$	0.8	211.9
$Li_{0.25}Mg_{0.5}MoS_2Cl_{0.5}$	0.75	244.9
$Li_{0.5}Mg_{0.5}MoS_2Cl_{0.5}$	0.67	277.4

Tables S4 Reported data on performance of different cathode materials.

Material	Electrolyte (mol)	Capacity ($mAhg^{-1}$) at 0.1C *	Voltage (V)	Morphology
Expanded MoS_2/graphene	0.25LiCl+0.25APC	300	1.2	Nanosheets[1]
Expanded TiS_2	0.2PY14Cl+0.25APC	239	0.7	Nanosheets[2]
TiS_2	$0.1Mg(BH_4)_2$ + $1.0NaBH_4$	200	~1.1	Nanosheets[3]
MoS_2	1LiCl+0.4APC	243	1.1	Nanoflakes[4]
TiO_2	$1.5LiBH_4$+$0.5Mg(BH_4)_2$/TG	140	1.15	Nanoparticles[5]
$Li_4Ti_5O_{12}$/Graphene	1.5LiBH4+0.4APC	147.5	~1.1	Micron-sized particles[6]
Mo_6S_8	0.4LiCl+1APC	126	~1.7	Particles[7]
$Ti_3C_2T_x$/CNT	0.4LiCl+0.5APC	105	-	Micron-sized delaminated flakes[8]

* Reported experimental data were based on active cathode (e.g. sulphide) materials in composite cathodes.

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Figure S1 Low energy structural configurations from USPEX global searching: (a) single layer MoS_2 (2H phase), (b) $Li_{0.5}MoS_2$, (c) $LiMoS_2$, (d) $Mg_{0.25}MoS_2$, (e) $Mg_{0.5}MoS_2$, (f) $Li_{0.5}Mg_{0.25}MoS_2$, and (g) $Li_{0.25}Mg_{0.5}MoS_2$. (b-g) exhibit characteristic layered structure of dT phases.

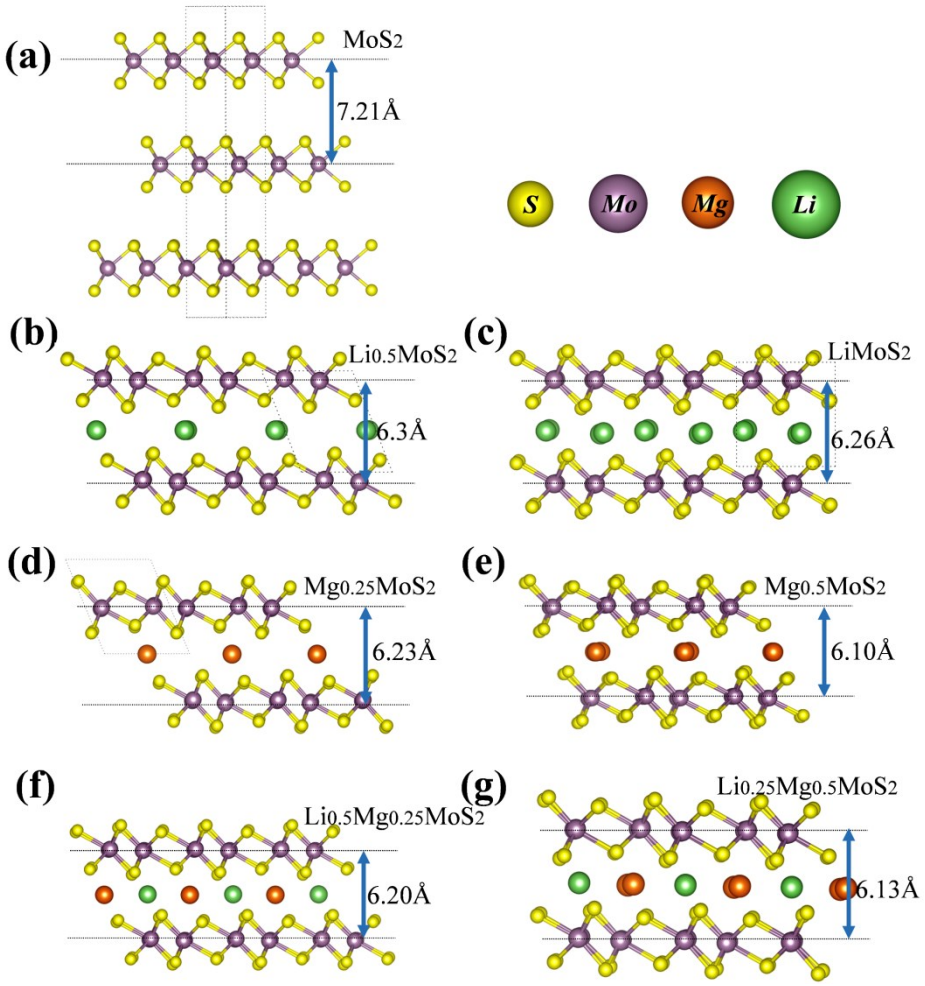

Figure S2 Low-energy structural configurations from USPEX global searching, for (a) $Li_{1.5}MoS_2$, (b) $Li_{0.5}Mg_{0.5}MoS_2$, and (c) $Mg_{0.75}MoS_2$. Higher level of intercalation of alkali and/or alkali-earth metal ions leads to loss of layered structures.

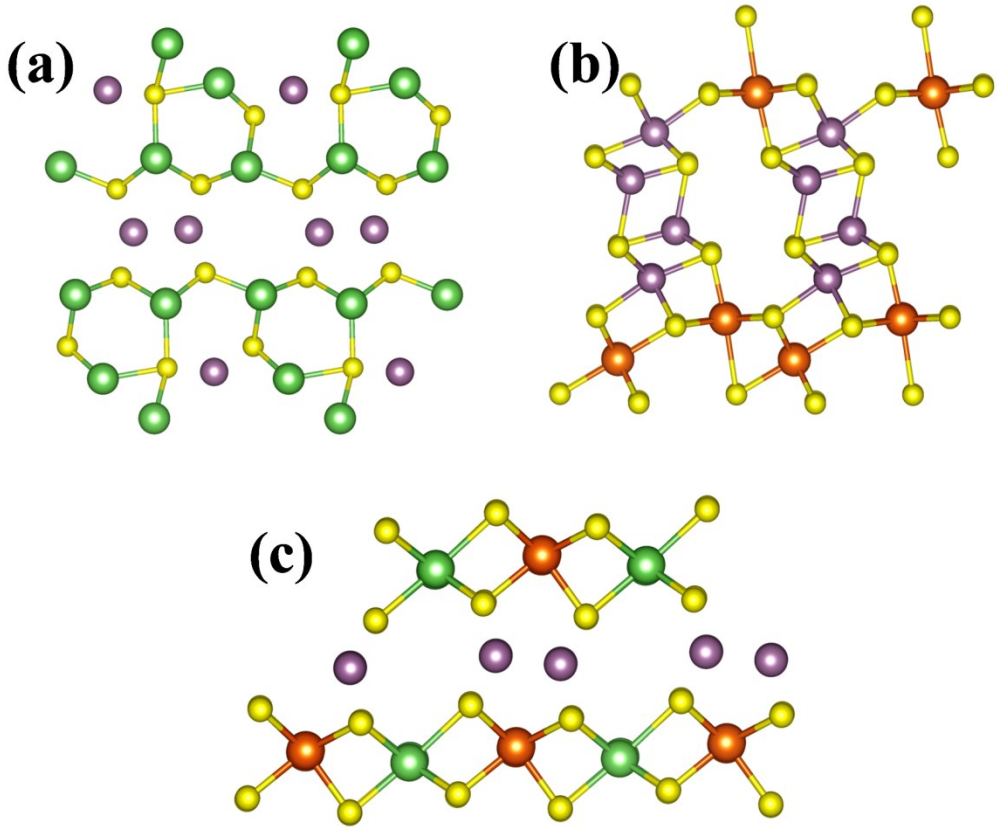

Figure S3 Reference AIMD simulation for the well-established LIB cathode $LiCoO_2$: (a) Layered structure of $LiCoO_2$, (b) Li^+ diffusion coefficient in $LiCoO_2$, and (c) trajectory map of Li^+ ions from the particle density distribution at 1200K.

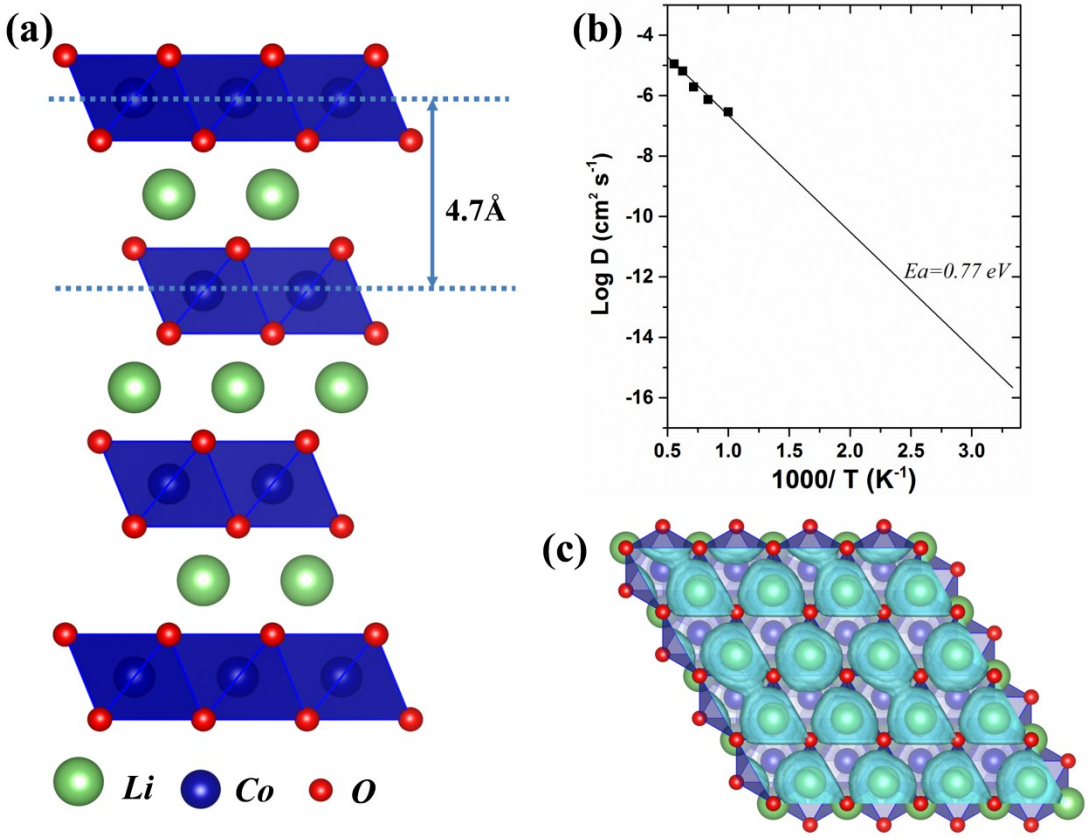

Figure S4 The trajectory of ions from AIMD at 1200K from the particle density distribution: Trajectories of (a) Cl^- and (b) Mg^{2+} in $Mg_{0.5}MoS_2Cl_{0.5}$; Trajectories of (c) Cl^-, (d) Mg^{2+}, and (e) Li^+ in $Li_{0.25}Mg_{0.25}MoS_2Cl_{0.5}$.