Supplementary Information

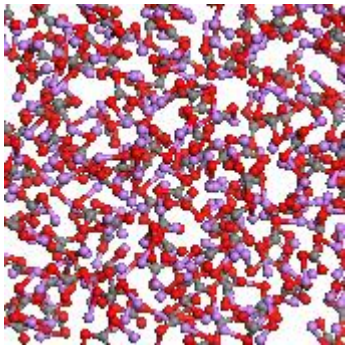
Understanding Lithium Transport in SEI Films: A Nonequilibrium Molecular Dynamics Simulation

Shaoyang Zhang[1], Yu Liu[*1] and Honglai Liu[2]

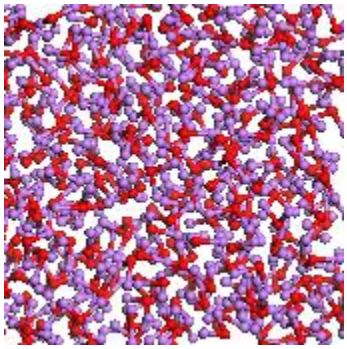
[1]*State Key Laboratory of Chemical Engineering and School of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, China*

[2]*State Key Laboratory of Chemical Engineering and School of Chemistry & Molecular Engineering, East China University of Science and Technology, Shanghai 200237, China*

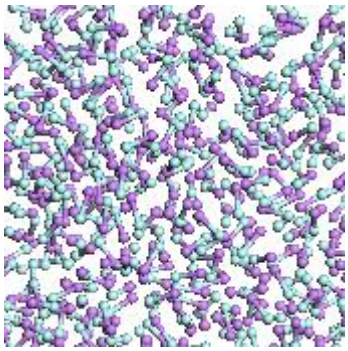
[*] Corresponding author liuyu89@mail.sysu.edu.cn



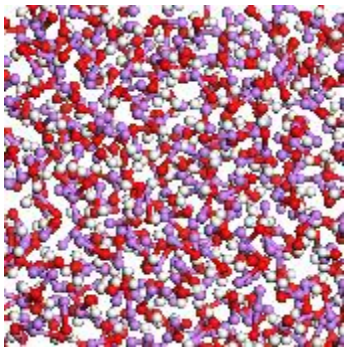
Li_2CO_3 ($1.5g/cm^3$)



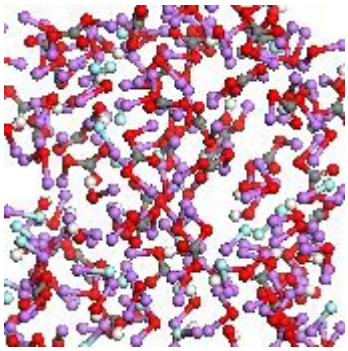
Li_2O ($1.5g/cm^3$)



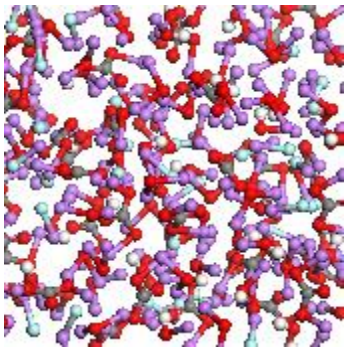
LiF ($1.5g/cm^3$)



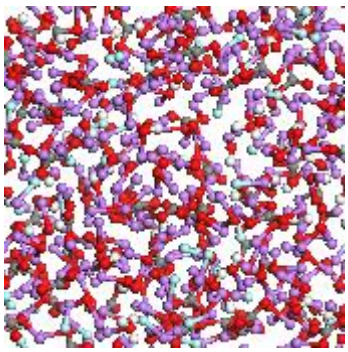
LiOH ($1.5g/cm^3$)



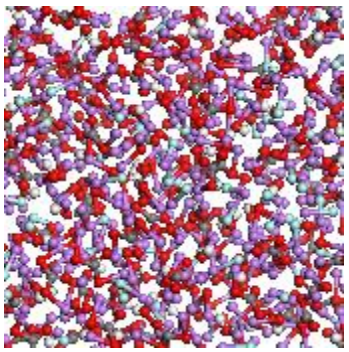
$Compound_1$ ($1g/cm^3$)



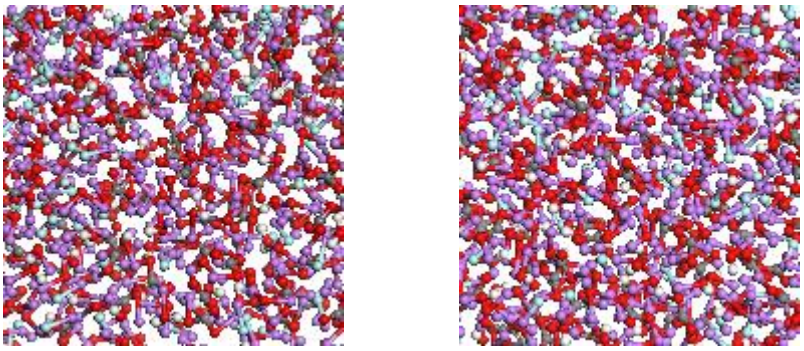
$Compound_{1.25}$ ($1.25g/cm^3$)



$Compound_{1.5}$ ($1.5g/cm^3$)

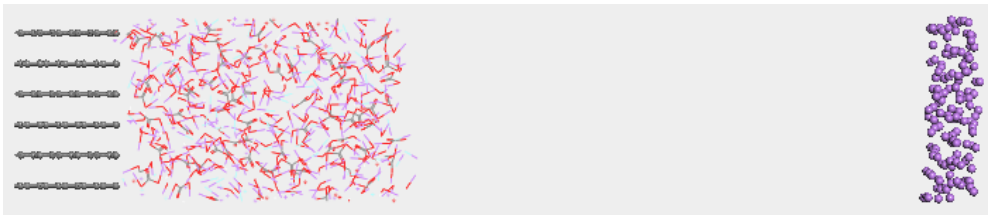


$Compound_{1.75}$ ($1.75g/cm^3$)

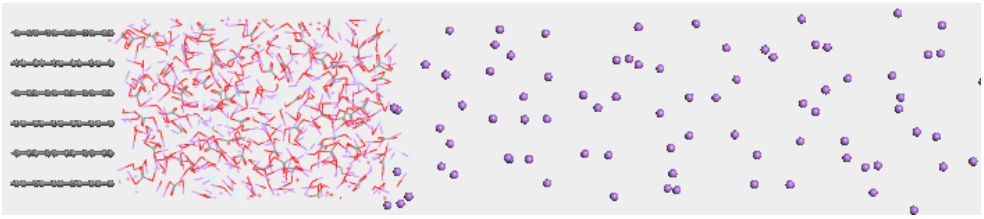


Compound$_{1.85}$ (1.85g/cm^3) Compound$_2$ (2g/cm^3)

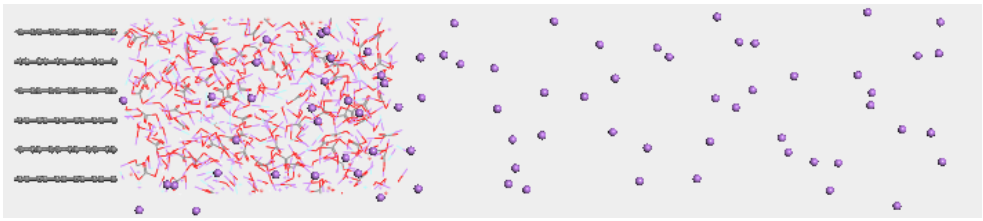
Figure S1. Molecular structures of the SEI film with different compositions: Li_2CO_3, Li_2O, LiF, LiOH and their compounds.



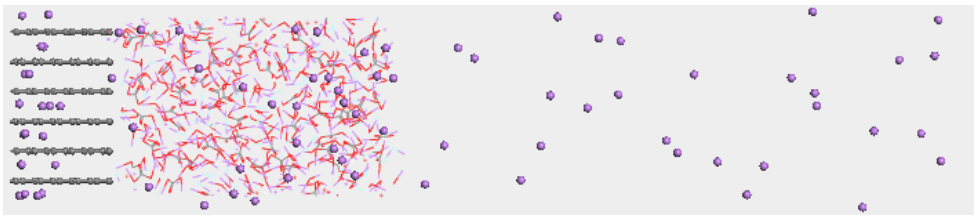
0 ps



1.6 ps



5.6 ps

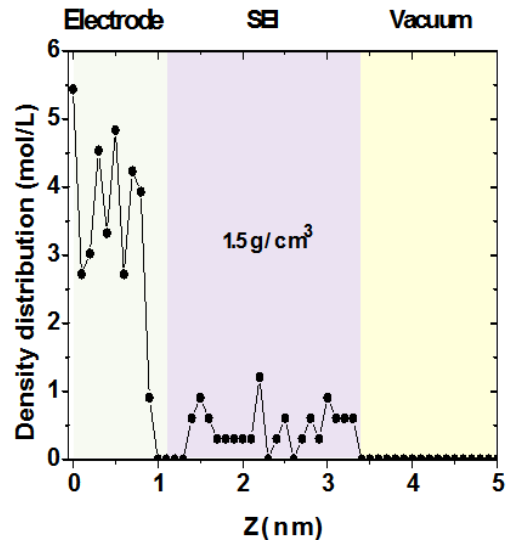


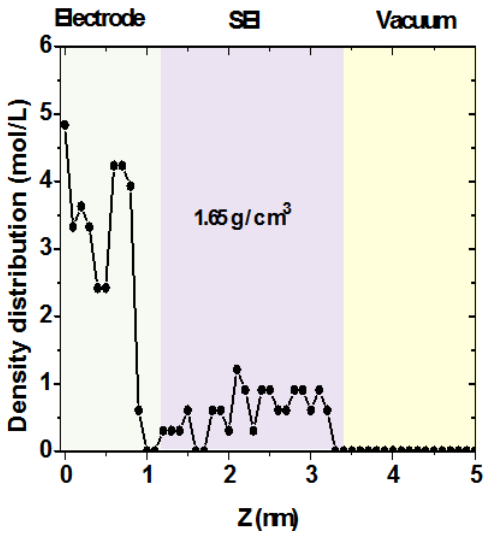
12 ps

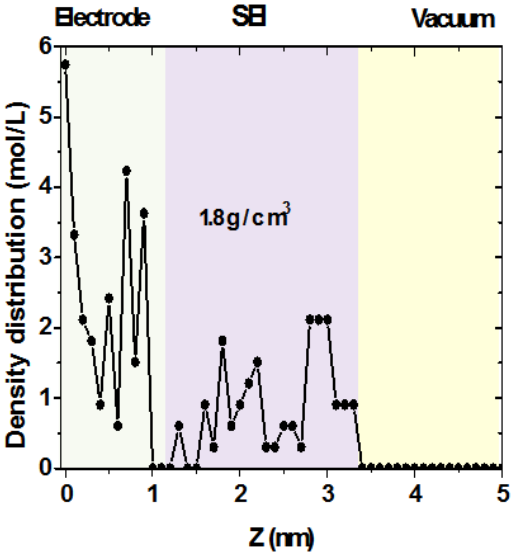
1000 ps

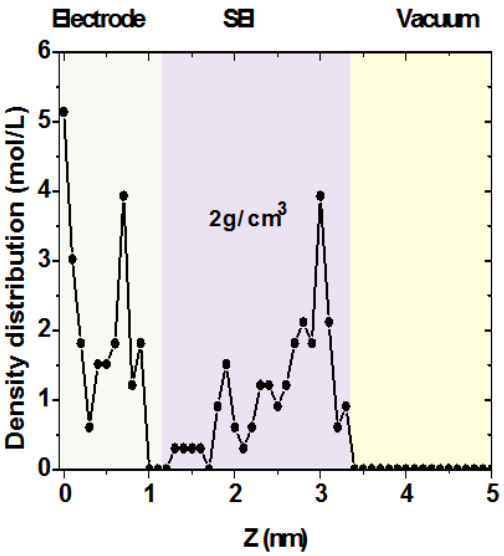
SEI Li C

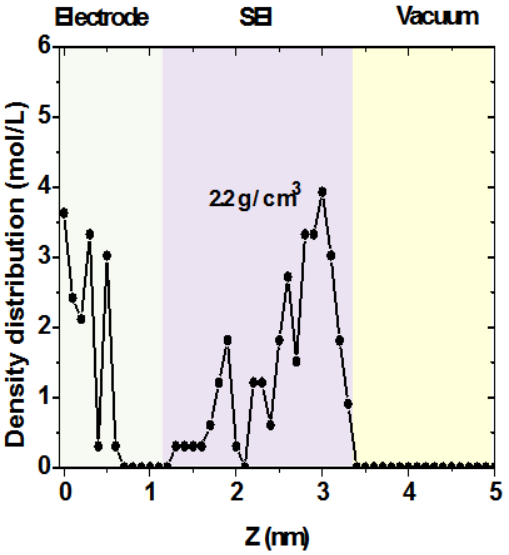
Figure S2. Snapshot of the transport process for compound$_{1.5}$.

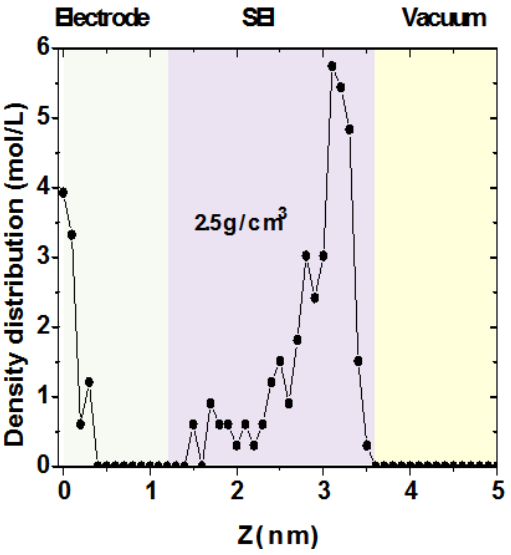

Electrode
SEI
Vacuum
Density distribution (mol/L)
1.5 g/cm3
Z (nm)


Electrode
SEI
Vacuum
Density distribution (mol/L)
1.65 g/cm3
Z (nm)


Electrode
SEI
Vacuum
Density distribution (mol/L)
1.8 g/cm3
Z (nm)


Electrode
SEI
Vacuum
Density distribution (mol/L)
2 g/cm3
Z (nm)


Electrode
SEI
Vacuum
Density distribution (mol/L)
2.2 g/cm3
Z (nm)


Electrode
SEI
Vacuum
Density distribution (mol/L)
2.5 g/cm3
Z (nm)

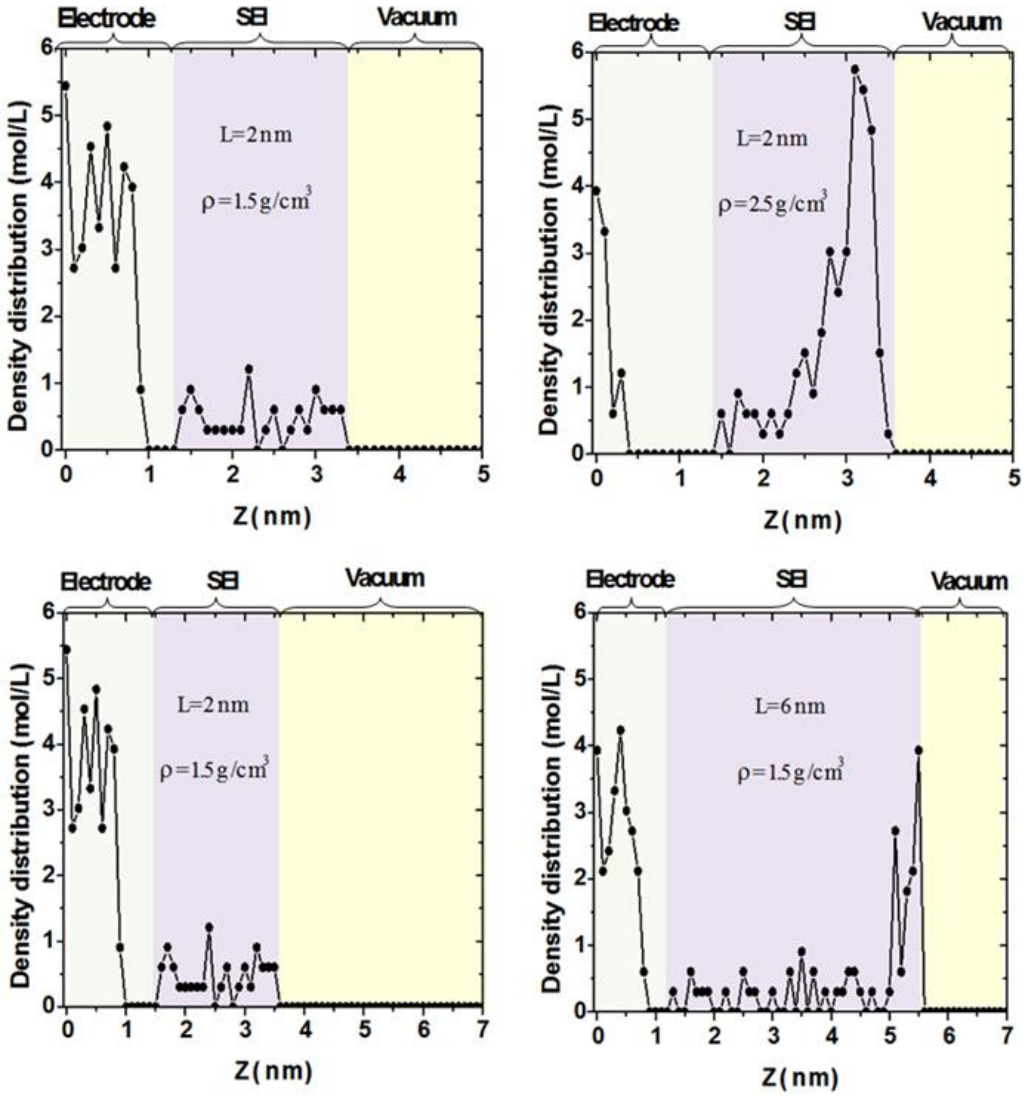


Figure S3. Density distribution of lithium ion in the SEI film with different film density when the system is in equilibrium; the composition is Li_2CO_3 : LiF : Li2O : LiOH = 1 : 1 : 1 : 1.

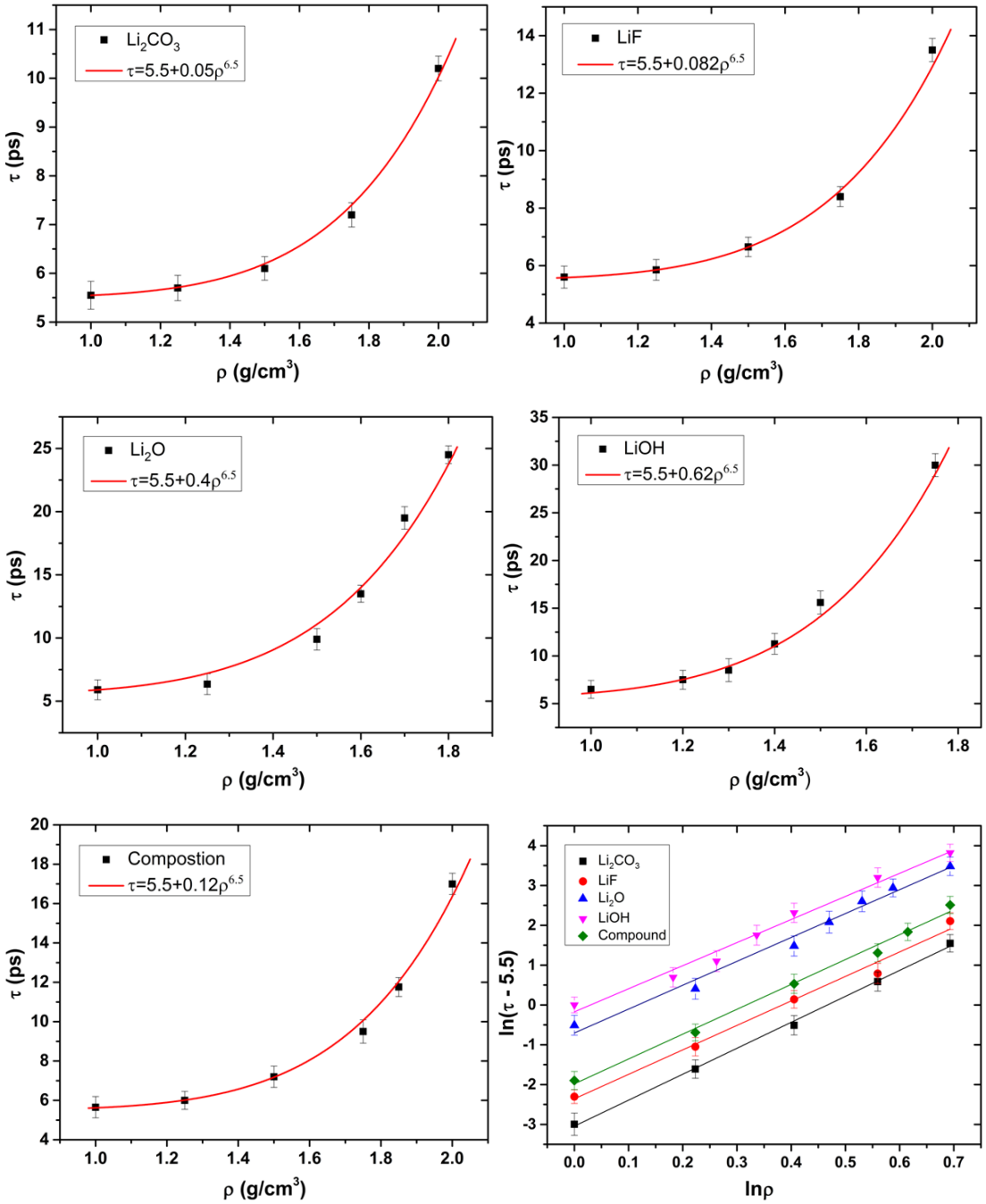


Figure S4. Data fitting of transition time of SEI films with different density, and the thickness of SEI film is 3 nm.

Table S1. Summary of the simulated and empirical values of different SEI films

$LiCO_3$:LiF:Li_2O:LiOH	L(nm)	ρ(g/cm^3)	S	$C(x_i)$	τ_{MD} (ps)	τ_{EMP} (ps)
1:1:1:1	1	1.00	1.2844	0.10	5.00	4.59
1:1:1:1	1	1.25	1.5207	0.10	5.05	4.79
1:1:1:1	1	1.50	1.8640	0.10	5.50	5.28
1:1:1:1	1	1.65	2.1491	0.10	5.80	5.91
1:1:1:1	1	1.80	2.5240	0.10	6.13	7.14
1:1:1:1	1	2.00	3.2424	0.10	8.40	10.24
1:1:1:1	2	1.00	1.6423	0.10	5.20	4.94
1:1:1:1	2	1.25	1.8785	0.10	5.45	5.31
1:1:1:1	2	1.50	2.2219	0.10	6.00	6.11
1:1:1:1	2	1.65	2.5070	0.10	6.82	7.07
1:1:1:1	2	1.80	2.8819	0.10	9.90	8.86
1:1:1:1	2	2.00	3.6003	0.10	13.45	14.41
1:1:1:1	3	1.00	1.9169	0.10	5.60	5.38
1:1:1:1	3	1.25	2.1532	0.10	6.00	5.92
1:1:1:1	3	1.50	2.4965	0.10	7.20	7.03
1:1:1:1	3	1.75	3.0194	0.10	9.20	9.68
1:1:1:1	3	1.85	3.3082	0.10	11.76	11.76
1:1:1:1	3	2.00	3.8749	0.10	18.10	17.45
1:1:1:1	4	1.00	2.1484	0.10	5.89	5.91
1:1:1:1	4	1.25	2.3847	0.10	6.65	6.62
1:1:1:1	4	1.50	2.7280	0.10	7.55	8.05
1:1:1:1	4	1.65	3.0131	0.10	8.75	9.64
1:1:1:1	4	1.80	3.3880	0.10	12.15	12.43
1:1:1:1	4	2.00	4.1064	0.10	20.00	20.50
1:1:1:1	5	1.00	2.3524	0.10	6.05	6.51
1:1:1:1	5	1.25	2.5886	0.10	6.68	7.41
1:1:1:1	5	1.50	2.9320	0.10	7.68	8.46
1:1:1:1	5	1.65	3.2171	0.10	9.55	10.52
1:1:1:1	5	1.80	3.5920	0.10	16.10	14.32
1:1:1:1	5	2.00	4.3104	0.10	22.00	23.58
1:1:1:1	6	1.00	2.5368	0.10	6.10	7.19
1:1:1:1	6	1.25	2.7730	0.10	6.60	8.27
1:1:1:1	6	1.50	3.1164	0.10	9.20	10.33
1:1:1:1	6	1.65	3.4015	0.10	10.25	12.54
1:1:1:1	6	1.80	3.7764	0.10	20.05	16.29
1:1:1:1	6	2.00	4.4948	0.10	27.00	26.71
1:1:1:1	7	1.00	2.7064	0.10	6.65	7.42
1:1:1:1	7	1.25	2.9426	0.10	7.35	9.21
1:1:1:1	7	1.50	3.2859	0.10	9.90	11.18
1:1:1:1	7	1.65	3.5710	0.10	13.70	14.11
1:1:1:1	7	1.80	3.9460	0.10	20.50	18.34
1:1:1:1	7	2.00	4.6643	0.10	30.00	29.91

$LiCO_3$:LiF:Li_2O:LiOH	L(nm)	ρ(g/cm^3)	S	$C(x_i)$	τ_{MD} (ps)	τ_{EMP} (ps)
2:1:1:1	2	1.50	2.2219	0.0953	5.85	6.03
3:1:1:1	2	1.50	2.2219	0.0868	5.70	5.88
4:1:1:1	2	1.50	2.2219	0.0755	5.50	5.68
1:2:1:1	2	1.50	2.2219	0.1003	5.94	6.12
1:3:1:1	2	1.50	2.2219	0.0953	5.85	6.03
1:4:1:1	2	1.50	2.2219	0.0896	5.75	5.93
1:1:2:1	2	1.50	2.2219	0.1122	6.15	6.33
1:1:3:1	2	1.50	2.2219	0.1280	6.43	6.61
1:1:4:1	2	1.50	2.2219	0.1355	6.56	6.74
1:1:1:2	2	1.50	2.2219	0.1239	6.18	6.53
1:1:1:3	2	1.50	2.2219	0.1413	6.49	6.84
1:1:1:4	2	1.50	2.2219	0.1780	7.14	7.49