

# Molecular Dynamics of Lithium Ion Transport in a Model Solid Electrolyte Interphase

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## Supporting Information

### Diffusion constant:

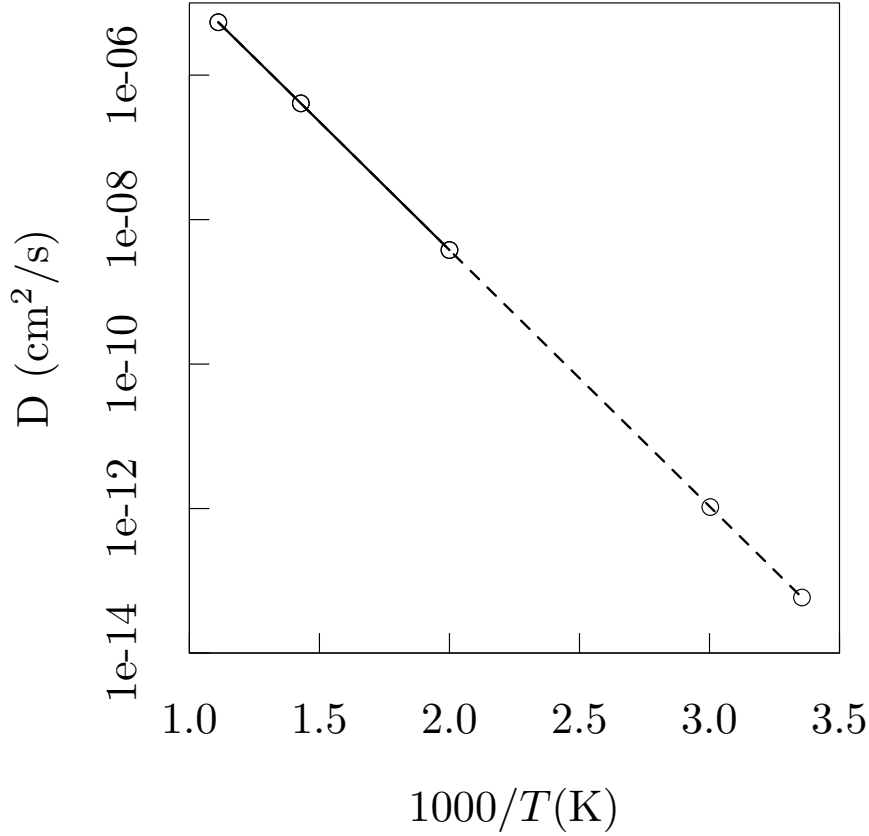


Figure 1: Arrhenius fit for the temperature dependence of  $\text{Li}^+$  diffusivity. The low temperature values ( $10^{-12}$  cm<sup>2</sup>/s at 333 K and  $5.8 \times 10^{-14}$  cm<sup>2</sup>/s at 298 K) are obtained by extrapolation.

### Conductivity:

At room temperature (298 K), the diffusion of EDC is insignificant compared to  $\text{Li}^+$  and hence only  $\text{Li}^+$  contributes to the conductivity. The conductivity ( $\lambda$ ) given by the Nernst-Einstein relation,  $\lambda = \frac{e^2}{V k_B T} (n_{\text{Li}} D_{\text{Li}})$ , is  $4.5 \times 10^{-9}$  S/cm.  $T$ ,  $V$ ,  $e$ ,  $k_B$ ,  $n_{\text{Li}}$ ,  $D_{\text{Li}}$  stand for

temperature, volume of the simulation box, electron charge, Boltzmann constant, number of  $\text{Li}^+$ , and diffusivity of  $\text{Li}^+$  respectively.

## Force Field Parameters:

```
; -----
; EDC topology for Solid Electrolyte Interphase simulations
; -----
; This is a standalone topology file
; Note:
; This might be a non-standard force field location. When you use this topology, the
; force field must either be present in the current directory, or the location
; specified in the GMXLIB path variable or with the 'include' mdp file option.
;
[ moleculetype ]
; Name          nrexcl
EDC              3

[ atoms ]
;  nr      type  resnr residue  atom  cgnr      charge      mass  typeB    chargeB
    1  op1s_465   1    EDC     C1    1   0.40600     12.011
    2  op1s_466   1    EDC     O1    1  -0.58600     15.9994
    3  op1s_466   1    EDC     O2    1  -0.58600     15.9994
    4  op1s_467   1    EDC     O3    1  -0.35600     15.9994
    5  op1s_468   1    EDC     C2    2   0.00800     12.011
    6  op1s_140   1    EDC     H1    2   0.05700      1.008
    7  op1s_140   1    EDC     H2    2   0.05700      1.008
    8  op1s_468   1    EDC     C3    3   0.00800     12.011
```

|    |          |   |     |    |   |          |         |
|----|----------|---|-----|----|---|----------|---------|
| 9  | opls_140 | 1 | EDC | H3 | 3 | 0.05700  | 1.008   |
| 10 | opls_140 | 1 | EDC | H4 | 3 | 0.05700  | 1.008   |
| 11 | opls_467 | 1 | EDC | O4 | 4 | -0.35600 | 15.9994 |
| 12 | opls_465 | 1 | EDC | C4 | 4 | 0.40600  | 12.011  |
| 13 | opls_466 | 1 | EDC | O5 | 4 | -0.58600 | 15.9994 |
| 14 | opls_466 | 1 | EDC | O6 | 4 | -0.58600 | 15.9994 |

[ bonds ]

|    |    |   |       |          |
|----|----|---|-------|----------|
| 1  | 2  | 1 | 0.127 | 476976.0 |
| 1  | 3  | 1 | 0.128 | 476976.0 |
| 1  | 4  | 1 | 0.148 | 179075.2 |
| 4  | 5  | 1 | 0.148 | 267776.0 |
| 5  | 6  | 1 | 0.110 | 284512.0 |
| 5  | 7  | 1 | 0.110 | 284512.0 |
| 5  | 8  | 1 | 0.153 | 224262.4 |
| 8  | 9  | 1 | 0.110 | 284512.0 |
| 8  | 10 | 1 | 0.110 | 284512.0 |
| 8  | 11 | 1 | 0.148 | 267776.0 |
| 11 | 12 | 1 | 0.148 | 179075.2 |
| 12 | 13 | 1 | 0.128 | 476976.0 |
| 12 | 14 | 1 | 0.127 | 476976.0 |

[ angles ]

|   |   |   |   |         |         |
|---|---|---|---|---------|---------|
| 1 | 4 | 5 | 1 | 117.162 | 694.544 |
| 2 | 1 | 3 | 1 | 131.306 | 200.000 |
| 2 | 1 | 4 | 1 | 112.723 | 694.544 |
| 3 | 1 | 4 | 1 | 115.971 | 694.544 |

|    |    |    |   |         |         |
|----|----|----|---|---------|---------|
| 4  | 5  | 6  | 1 | 110.294 | 292.880 |
| 4  | 5  | 7  | 1 | 110.284 | 292.880 |
| 4  | 5  | 8  | 1 | 106.028 | 418.400 |
| 5  | 8  | 9  | 1 | 111.075 | 313.800 |
| 5  | 8  | 10 | 1 | 111.050 | 313.800 |
| 5  | 8  | 11 | 1 | 106.028 | 418.400 |
| 6  | 5  | 7  | 1 | 108.123 | 276.144 |
| 6  | 5  | 8  | 1 | 111.050 | 313.800 |
| 7  | 5  | 8  | 1 | 111.075 | 313.800 |
| 8  | 11 | 12 | 1 | 117.162 | 694.544 |
| 9  | 8  | 10 | 1 | 108.123 | 276.144 |
| 9  | 8  | 11 | 1 | 110.284 | 292.880 |
| 10 | 8  | 11 | 1 | 110.294 | 292.880 |
| 11 | 12 | 13 | 1 | 115.971 | 694.544 |
| 11 | 12 | 14 | 1 | 112.723 | 694.544 |
| 13 | 12 | 14 | 1 | 131.306 | 200.000 |

[ dihedrals ]

|   |   |   |    |   |
|---|---|---|----|---|
| 2 | 1 | 4 | 5  | 3 |
| 3 | 1 | 4 | 5  | 3 |
| 1 | 4 | 5 | 6  | 3 |
| 1 | 4 | 5 | 7  | 3 |
| 1 | 4 | 5 | 8  | 3 |
| 4 | 5 | 8 | 9  | 3 |
| 6 | 5 | 8 | 9  | 3 |
| 7 | 5 | 8 | 9  | 3 |
| 4 | 5 | 8 | 10 | 3 |

6 5 8 10 3  
7 5 8 10 3  
4 5 8 11 3  
6 5 8 11 3  
7 5 8 11 3  
5 8 11 12 3  
9 8 11 12 3  
10 8 11 12 3  
8 11 12 13 3  
8 11 12 14 3

[ pairs ]

5 2 1  
5 3 1  
6 1 1  
7 1 1  
8 1 1  
9 4 1  
9 6 1  
9 7 1  
10 4 1  
10 6 1  
10 7 1  
11 4 1  
11 6 1  
11 7 1  
12 5 1

```

12 9 1
12 10 1
13 8 1
14 8 1

; -----
; EC solvent topology for battery electrolytes
; -----
; Charges on EC atoms scaled to 80%
; This is a standalone topology file
;
; Note:
; This might be a non-standard force field location. When you use this topology, the
; force field must either be present in the current directory, or the location
; specified in the GMXLIB path variable or with the 'include' mdp file option.
;

[ moleculetype ]
; Name          nrexcl
EC              3

[ atoms ]
;  nr      type  resnr residue  atom   cgnr    charge      mass  typeB    chargeB
; residue  0 EC  rtp EC    q  0.0
      1  opls_774      0    EC    C3      1      0.0264   12.011  ; qtot 0.21
      2  opls_777      0    EC    H5      1      0.08328   1.008  ; qtot 0.24
      3  opls_777      0    EC    H6      1      0.08328   1.008  ; qtot 0.27
      4  opls_773      0    EC    O1      2     -0.37472  15.9994 ; qtot -0.18

```

|    |          |   |    |    |   |          |         |               |
|----|----------|---|----|----|---|----------|---------|---------------|
| 5  | opls_772 | 0 | EC | C4 | 2 | 0.87968  | 12.011  | ; qtot 0.68   |
| 6  | opls_771 | 0 | EC | O2 | 2 | -0.51616 | 15.9994 | ; qtot 0.18   |
| 7  | opls_773 | 0 | EC | O3 | 2 | -0.37472 | 15.9994 | ; qtot -0.27  |
| 8  | opls_774 | 0 | EC | C2 | 3 | 0.0264   | 12.011  | ; qtot -0.11  |
| 9  | opls_777 | 0 | EC | H4 | 3 | 0.08328  | 1.008   | ; qtot -0.055 |
| 10 | opls_777 | 0 | EC | H3 | 3 | 0.08328  | 1.008   | ; qtot 0      |

[ bonds ]

| ; ai | aj | funct | c0 | c1 | c2 | c3 |
|------|----|-------|----|----|----|----|
| 1    | 2  | 1     |    |    |    |    |
| 1    | 3  | 1     |    |    |    |    |
| 1    | 4  | 1     |    |    |    |    |
| 1    | 8  | 1     |    |    |    |    |
| 4    | 5  | 1     |    |    |    |    |
| 5    | 6  | 1     |    |    |    |    |
| 5    | 7  | 1     |    |    |    |    |
| 7    | 8  | 1     |    |    |    |    |
| 8    | 9  | 1     |    |    |    |    |
| 8    | 10 | 1     |    |    |    |    |

[ pairs ]

| ; ai | aj | funct | c0 | c1 | c2 | c3 |
|------|----|-------|----|----|----|----|
| 1    | 6  | 1     |    |    |    |    |
| 2    | 5  | 1     |    |    |    |    |
| 2    | 7  | 1     |    |    |    |    |
| 2    | 9  | 1     |    |    |    |    |
| 2    | 10 | 1     |    |    |    |    |

|   |    |   |
|---|----|---|
| 3 | 5  | 1 |
| 3 | 7  | 1 |
| 3 | 9  | 1 |
| 3 | 10 | 1 |
| 4 | 9  | 1 |
| 4 | 10 | 1 |
| 5 | 9  | 1 |
| 5 | 10 | 1 |
| 6 | 8  | 1 |

[ angles ]

| ; | ai | aj | ak | funct | c0 | c1 | c2 | c3 |
|---|----|----|----|-------|----|----|----|----|
|   | 2  | 1  | 3  | 1     |    |    |    |    |
|   | 2  | 1  | 4  | 1     |    |    |    |    |
|   | 2  | 1  | 8  | 1     |    |    |    |    |
|   | 3  | 1  | 4  | 1     |    |    |    |    |
|   | 3  | 1  | 8  | 1     |    |    |    |    |
|   | 4  | 1  | 8  | 1     |    |    |    |    |
|   | 1  | 4  | 5  | 1     |    |    |    |    |
|   | 4  | 5  | 6  | 1     |    |    |    |    |
|   | 6  | 5  | 7  | 1     |    |    |    |    |
|   | 5  | 7  | 8  | 1     |    |    |    |    |
|   | 1  | 8  | 7  | 1     |    |    |    |    |
|   | 1  | 8  | 9  | 1     |    |    |    |    |
|   | 1  | 8  | 10 | 1     |    |    |    |    |
|   | 7  | 8  | 9  | 1     |    |    |    |    |
|   | 7  | 8  | 10 | 1     |    |    |    |    |

```

      9      8      10      1

[ dihedrals ]

; ai      aj      ak      al funct      c0      c1      c2      c3
  2       1       4       5       3
  3       1       4       5       3
  8       1       4       5       3
  2       1       8       7       3
  2       1       8       9       3
  2       1       8      10       3
  3       1       8       7       3
  3       1       8       9       3
  3       1       8      10       3
  4       1       8       7       3
  4       1       8       9       3
  4       1       8      10       3
  1       4       5       6       3
  6       5       7       8       3
  5       7       8       1       3
  5       7       8       9       3
  5       7       8      10       3

; -----

```