

# Supporting Information

## Halogen-doping effect on the delithiation and charge transfer of $(\text{Li}_2\text{S})_{10}$ cluster

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## S1. Additional Computational details

As described in the Computational Methods section the simulation of the delithiation (charge) process in the halogen-doped structures was performed based on the methodology reported by Liu et. al through ab initio molecular dynamics simulations followed by DFT calculations.<sup>20</sup>

The followed methodology to select the delithiation sites as explained in reference 20, is selecting the four lithium atoms farthest from the center of mass of the cluster, among those Li-atoms chosen it was included one close to the halogen atom. Then select two of these four atoms and remove them, ending with six different combinations which result in six  $\text{Li}_{20-x} \text{S}_{10}\text{I}$  structures with different configurations.

Then each structure is relaxed through AIMD simulation at 300K for 2 ps with a time step of 2 fs, and then a PAW-PBE geometry optimization is performed. Thus, the system with the lowest energy is selected and used as starting point for exploring the next delithiation step.

## S2. Coordinates of the calculated molecular geometries

The coordinates of the optimized structures for each delithiation step are presented next in the VASP (POSCAR) format.

$\text{Li}_{20}\text{S}_{10}\text{I}$

1.0

|               |               |               |
|---------------|---------------|---------------|
| 20.0000000000 | 0.0000000000  | 0.0000000000  |
| 0.0000000000  | 20.0000000000 | 0.0000000000  |
| 0.0000000000  | 0.0000000000  | 20.0000000000 |

Li S I

20 10 1

Direct

|             |             |             |
|-------------|-------------|-------------|
| 0.652455378 | 0.485463428 | 0.469210052 |
| 0.510356522 | 0.469326401 | 0.315647340 |
| 0.374132347 | 0.358959174 | 0.444320297 |
| 0.481746960 | 0.432962513 | 0.478326941 |
| 0.453668165 | 0.459759808 | 0.627933407 |
| 0.306798434 | 0.553130054 | 0.346792078 |
| 0.507421541 | 0.316930079 | 0.476274347 |

|             |             |             |
|-------------|-------------|-------------|
| 0.535835838 | 0.362870717 | 0.605042696 |
| 0.631629086 | 0.611118889 | 0.437117958 |
| 0.390445900 | 0.447021532 | 0.347666788 |
| 0.586173725 | 0.399483562 | 0.394170094 |
| 0.501239729 | 0.552584124 | 0.422102165 |
| 0.356923962 | 0.454483795 | 0.530698681 |
| 0.368052721 | 0.560200500 | 0.452855539 |
| 0.533612919 | 0.627327251 | 0.528332806 |
| 0.542811775 | 0.605009317 | 0.319836116 |
| 0.609979677 | 0.557031679 | 0.660409737 |
| 0.425340462 | 0.664975357 | 0.374586821 |
| 0.652996588 | 0.440709782 | 0.601219893 |
| 0.555830479 | 0.494412851 | 0.559472513 |
| 0.473694134 | 0.370180702 | 0.374104691 |
| 0.307499981 | 0.459377861 | 0.425491333 |
| 0.424760294 | 0.359647369 | 0.556547880 |
| 0.561535263 | 0.454821587 | 0.673899126 |
| 0.602826214 | 0.385512638 | 0.508858109 |
| 0.606998444 | 0.514414406 | 0.365388489 |
| 0.428642321 | 0.555663013 | 0.323311090 |
| 0.450982761 | 0.536395788 | 0.531253290 |
| 0.533848476 | 0.671541977 | 0.418077850 |
| 0.644033670 | 0.570495319 | 0.550935078 |
| 0.297356153 | 0.670818424 | 0.414396858 |

1<sup>st</sup> delithiation

Li<sub>18</sub>S<sub>10</sub>I

1.0

20.000000000000 0.000000000000 0.000000000000

0.000000000000 20.000000000000 0.000000000000

0.000000000000 0.000000000000 20.000000000000

Li S I

18 10 1

Direct

0.645482544358 0.554015331461 0.521479072528  
0.426982995733 0.406765635649 0.345779085216  
0.446132961733 0.359715749524 0.472079280216  
0.461732979545 0.492615647336 0.429829562091  
0.454582905358 0.439465723774 0.675129344466  
0.570582915920 0.363265697711 0.509429531091  
0.584232865483 0.385165407836 0.644629157278  
0.592932641795 0.666965587399 0.487179104466  
0.338932554920 0.514815367961 0.362179307091  
0.621582588170 0.458965349899 0.439979213091  
0.543082833733 0.492165631586 0.326429464341  
0.390182588045 0.428265507836 0.559879360153  
0.349382744608 0.539765360899 0.507629468091  
0.482832485608 0.610315365274 0.544129337091  
0.439282898045 0.613615480774 0.335279245903  
0.510582928670 0.574115506774 0.672229595091  
0.635982563983 0.524415703961 0.647229424341  
0.525682752920 0.476665441211 0.574379163403  
0.533832686045 0.397165299649 0.400229406403  
0.357732860795 0.435815419961 0.447029459841  
0.481132778233 0.359165378149 0.585829212028  
0.556932966358 0.474365712836 0.714629318466  
0.644332680170 0.443965353086 0.554979364716  
0.614532869608 0.576615418211 0.404629208278  
0.432682667045 0.508665680899 0.292079318466  
0.418632879545 0.533465668586 0.603329120278

|                |                |                |
|----------------|----------------|----------------|
| 0.513382850608 | 0.603365474149 | 0.426579096216 |
| 0.589032505295 | 0.623715482149 | 0.594029322966 |
| 0.337582507670 | 0.642615615461 | 0.421779456403 |

2<sup>nd</sup> delithiation

Li<sub>16</sub>S<sub>10</sub>I

1.0

|                 |                 |                 |
|-----------------|-----------------|-----------------|
| 20.000000000000 | 0.000000000000  | 0.000000000000  |
| 0.000000000000  | 20.000000000000 | 0.000000000000  |
| 0.000000000000  | 0.000000000000  | 20.000000000000 |

Li S I

16 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.632250266000 | 0.555525140000 | 0.495949802000 |
| 0.431399804000 | 0.343074794000 | 0.484099880000 |
| 0.508349912000 | 0.438424880000 | 0.480149906000 |
| 0.448100150000 | 0.465825182000 | 0.678699914000 |
| 0.560000246000 | 0.337924742000 | 0.532599944000 |
| 0.569399900000 | 0.412925180000 | 0.643800038000 |
| 0.552149888000 | 0.656425130000 | 0.454499870000 |
| 0.623949944000 | 0.400375256000 | 0.429599948000 |
| 0.461749928000 | 0.400525070000 | 0.357250226000 |
| 0.387849866000 | 0.450375026000 | 0.559349834000 |
| 0.367749734000 | 0.557425220000 | 0.495749876000 |
| 0.456599876000 | 0.643224794000 | 0.554150192000 |
| 0.437850158000 | 0.540074984000 | 0.374549828000 |
| 0.503149748000 | 0.599474930000 | 0.670650152000 |
| 0.628650032000 | 0.541174838000 | 0.630899852000 |
| 0.516999974000 | 0.517224956000 | 0.581249822000 |
| 0.531000014000 | 0.333625028000 | 0.420899774000 |

|                |                |                |
|----------------|----------------|----------------|
| 0.393249956000 | 0.456025154000 | 0.441199832000 |
| 0.466500128000 | 0.374925146000 | 0.594999824000 |
| 0.553349966000 | 0.504675032000 | 0.708800000000 |
| 0.620899898000 | 0.448575170000 | 0.538700036000 |
| 0.622049864000 | 0.589924940000 | 0.379600178000 |
| 0.511000106000 | 0.495875156000 | 0.291200000000 |
| 0.402949760000 | 0.556425068000 | 0.608299862000 |
| 0.569750162000 | 0.502474802000 | 0.373700012000 |
| 0.571299980000 | 0.628175012000 | 0.566000114000 |
| 0.418199990000 | 0.666374972000 | 0.424249970000 |

3<sup>rd</sup> delithiation

Li<sub>14</sub>S<sub>10</sub>I

1.0

|                  |                  |                  |
|------------------|------------------|------------------|
| 20.0000000000000 | 0.0000000000000  | 0.0000000000000  |
| 0.0000000000000  | 20.0000000000000 | 0.0000000000000  |
| 0.0000000000000  | 0.0000000000000  | 20.0000000000000 |

Li S I

14 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.629475005875 | 0.472624890375 | 0.540200098000 |
| 0.370274988750 | 0.469774733312 | 0.573299947563 |
| 0.487675022375 | 0.436625074250 | 0.508000172000 |
| 0.463125165062 | 0.382524773188 | 0.635600137563 |
| 0.592725173625 | 0.590375027312 | 0.478450207188 |
| 0.523975132312 | 0.308425000000 | 0.514499832813 |
| 0.403175121375 | 0.441724800750 | 0.403150216625 |
| 0.377224850812 | 0.593724716188 | 0.672450067750 |
| 0.406825104062 | 0.647374816000 | 0.507949883563 |
| 0.538175150250 | 0.681225160625 | 0.568450226438 |

|                |                |                |
|----------------|----------------|----------------|
| 0.473624911875 | 0.567024908437 | 0.391549871437 |
| 0.509874733375 | 0.603474925813 | 0.676700159125 |
| 0.552025064875 | 0.478024910687 | 0.648300123250 |
| 0.471925162687 | 0.560724964562 | 0.574599783938 |
| 0.427425163812 | 0.339874909875 | 0.457399946188 |
| 0.409174771437 | 0.530174978250 | 0.480100146875 |
| 0.382774778562 | 0.352324890188 | 0.550900040687 |
| 0.436625074250 | 0.495925199750 | 0.672249871875 |
| 0.568225125812 | 0.384524816188 | 0.583249874125 |
| 0.629725011250 | 0.494325069563 | 0.418250158125 |
| 0.492825037312 | 0.469825021750 | 0.323299840875 |
| 0.434725129187 | 0.673274798125 | 0.617900044375 |
| 0.555124748375 | 0.429274820438 | 0.392399985500 |
| 0.592274972375 | 0.576324916813 | 0.592949795313 |
| 0.519674752500 | 0.691575000000 | 0.427450068563 |

#### 4<sup>th</sup> delithiation

Li<sub>12</sub>S<sub>10</sub>I

1.0

|                 |                 |                 |
|-----------------|-----------------|-----------------|
| 20.000000000000 | 0.000000000000  | 0.000000000000  |
| 0.000000000000  | 20.000000000000 | 0.000000000000  |
| 0.000000000000  | 0.000000000000  | 20.000000000000 |

Li S I

12 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.627899881250 | 0.471949838125 | 0.505724812500 |
| 0.376500181250 | 0.539549938125 | 0.618074967500 |
| 0.499450228750 | 0.421449895000 | 0.479924833125 |
| 0.453900119375 | 0.431850128750 | 0.606324908125 |
| 0.587750020625 | 0.607449999375 | 0.441375081250 |

|                |                |                |
|----------------|----------------|----------------|
| 0.504400062500 | 0.310750000000 | 0.565425144375 |
| 0.372100118750 | 0.422549910625 | 0.431774901875 |
| 0.409800138125 | 0.622000012500 | 0.520975050625 |
| 0.450450091875 | 0.549549908125 | 0.406375186250 |
| 0.547499857500 | 0.663600001250 | 0.574625060000 |
| 0.593750191875 | 0.506699923125 | 0.637375155625 |
| 0.511099985625 | 0.533350108125 | 0.554625120000 |
| 0.423199984375 | 0.323150133125 | 0.478974798125 |
| 0.404099928125 | 0.506199830000 | 0.508574917500 |
| 0.388350070000 | 0.331900106875 | 0.575925207500 |
| 0.478349800000 | 0.538500073750 | 0.674725062500 |
| 0.565250088125 | 0.407750087500 | 0.577625145625 |
| 0.562549963750 | 0.492200061250 | 0.400974937500 |
| 0.412399960000 | 0.468450132500 | 0.325274937500 |
| 0.448450192500 | 0.632749885625 | 0.634375070000 |
| 0.499450228750 | 0.423499945625 | 0.352725176875 |
| 0.621550070625 | 0.581449888125 | 0.546874859375 |
| 0.481400036875 | 0.689250000000 | 0.431975033750 |

### 5<sup>th</sup> delithiation

Li<sub>10</sub>S<sub>10</sub>I  
 1.0  
 20.0000000000000 0.000000000000 0.000000000000  
 0.000000000000 20.000000000000 0.000000000000  
 0.000000000000 0.000000000000 20.000000000000

### Li S I

10 10 1

### Direct

0.597600152500 0.508099893750 0.400250022500

|                |                |                |
|----------------|----------------|----------------|
| 0.446150213125 | 0.445199914375 | 0.623750015000 |
| 0.535200055000 | 0.370200035000 | 0.403949909375 |
| 0.550499960000 | 0.363400024375 | 0.593849849375 |
| 0.547100192500 | 0.559650033750 | 0.536649760000 |
| 0.349700121875 | 0.389599827500 | 0.528800045000 |
| 0.426950183125 | 0.578750182500 | 0.617200183125 |
| 0.390150125625 | 0.584750191875 | 0.478600204375 |
| 0.626400197500 | 0.484700095000 | 0.618350244375 |
| 0.456799932500 | 0.460099818750 | 0.481699852500 |
| 0.432950192500 | 0.340749810625 | 0.458499816250 |
| 0.370399797500 | 0.499500118125 | 0.554050025000 |
| 0.444349972500 | 0.323350021250 | 0.560800095000 |
| 0.545600071250 | 0.434499778750 | 0.690250000000 |
| 0.575349939375 | 0.429699771250 | 0.500949823125 |
| 0.483000211250 | 0.554700204375 | 0.404650029375 |
| 0.558050031250 | 0.437949962500 | 0.309750000000 |
| 0.530750107500 | 0.535449758125 | 0.659650190000 |
| 0.467950009375 | 0.470149775000 | 0.346150056875 |
| 0.650299878125 | 0.504999770000 | 0.504849948125 |
| 0.471900075000 | 0.676649978750 | 0.535600055625 |

#### 6<sup>th</sup> delithiation

Li<sub>8</sub>S<sub>10</sub> I  
 1.0  
 20.0000000000000 0.000000000000 0.000000000000  
 0.000000000000 20.000000000000 0.000000000000  
 0.000000000000 0.000000000000 20.000000000000

#### Li S I

8 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.523775082500 | 0.622950100000 | 0.411175168125 |
| 0.466725143125 | 0.460049846250 | 0.641224833750 |
| 0.523275094375 | 0.405299948125 | 0.525524801250 |
| 0.530375117500 | 0.557549927500 | 0.549924988750 |
| 0.386075093125 | 0.398899812500 | 0.496525010625 |
| 0.398074808125 | 0.597950214375 | 0.499374895000 |
| 0.648225146875 | 0.473000161875 | 0.532074981250 |
| 0.485324893125 | 0.490000237500 | 0.428675231875 |
| 0.452124818750 | 0.308250000000 | 0.536775253125 |
| 0.430675184375 | 0.495549961875 | 0.539024960000 |
| 0.476025018125 | 0.335850015625 | 0.633574967500 |
| 0.575825140625 | 0.365199750000 | 0.640724845625 |
| 0.593025115625 | 0.439650043125 | 0.434774799375 |
| 0.407324828125 | 0.588400105625 | 0.376825073125 |
| 0.401275115625 | 0.419800083125 | 0.371474768750 |
| 0.588124944375 | 0.470550076250 | 0.635825153750 |
| 0.351774853125 | 0.505199780625 | 0.358775166250 |
| 0.600125138750 | 0.544649946250 | 0.452724996250 |
| 0.492124827500 | 0.691750000000 | 0.521024908125 |

### 7<sup>th</sup> delithiation

Li<sub>6</sub>S<sub>10</sub> I  
 1.0  
 20.0000000000000 0.000000000000 0.000000000000  
 0.000000000000 20.000000000000 0.000000000000  
 0.000000000000 0.000000000000 20.000000000000

### Li S I

6 10 1

### Direct

0.570424910000 0.590799857000 0.440800124000

|                |                |                |
|----------------|----------------|----------------|
| 0.477724968500 | 0.408200121500 | 0.508850037500 |
| 0.484525202000 | 0.537349995500 | 0.551000223500 |
| 0.410275242500 | 0.610950006500 | 0.443949908000 |
| 0.613974951500 | 0.384100083500 | 0.548200076000 |
| 0.502075121000 | 0.472449866000 | 0.381199977500 |
| 0.522574907000 | 0.331500087500 | 0.605049836000 |
| 0.397675233500 | 0.492549818000 | 0.457350021500 |
| 0.556525004000 | 0.397250082500 | 0.670699872500 |
| 0.483825056000 | 0.479150141000 | 0.674600000000 |
| 0.575025183500 | 0.403049858000 | 0.439500227000 |
| 0.534575165000 | 0.575300178500 | 0.328349867000 |
| 0.386025048500 | 0.502049804000 | 0.351999873500 |
| 0.413575182500 | 0.457099907000 | 0.608650088000 |
| 0.432524957000 | 0.591300086000 | 0.325400000000 |
| 0.585975222500 | 0.488049939500 | 0.503100023000 |
| 0.497325128000 | 0.668499912500 | 0.525049862000 |

8<sup>th</sup> delithiation

Li<sub>4</sub>S<sub>10</sub> I

1.0

|                 |                 |                 |
|-----------------|-----------------|-----------------|
| 20.000000000000 | 0.000000000000  | 0.000000000000  |
| 0.000000000000  | 20.000000000000 | 0.000000000000  |
| 0.000000000000  | 0.000000000000  | 20.000000000000 |

Li S I

4 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.565175049375 | 0.523325056937 | 0.400225029125 |
| 0.482774764438 | 0.453075173875 | 0.502224910063 |
| 0.570074948375 | 0.547425009750 | 0.574475095688 |
| 0.441275073312 | 0.599625088563 | 0.521224976813 |

|                |                |                |
|----------------|----------------|----------------|
| 0.540324821125 | 0.357825180250 | 0.569575196688 |
| 0.371074863063 | 0.515075050687 | 0.471074873375 |
| 0.540275148313 | 0.387075123875 | 0.668525166938 |
| 0.527275125375 | 0.491074875437 | 0.672775000000 |
| 0.628925136938 | 0.380324804625 | 0.521725160438 |
| 0.487275121250 | 0.434124779938 | 0.377674868063 |
| 0.380124817563 | 0.564524983437 | 0.385724887250 |
| 0.452175016125 | 0.512124917562 | 0.605124948750 |
| 0.456375176375 | 0.513724814062 | 0.327225000000 |
| 0.621624961250 | 0.479675180938 | 0.495424918000 |
| 0.555774793563 | 0.642174819750 | 0.471924926375 |

#### 9<sup>th</sup> delithiation

Li<sub>2</sub>S<sub>10</sub> I

1.0

|                 |                 |                 |
|-----------------|-----------------|-----------------|
| 20.000000000000 | 0.000000000000  | 0.000000000000  |
| 0.000000000000  | 20.000000000000 | 0.000000000000  |
| 0.000000000000  | 0.000000000000  | 20.000000000000 |

Li S I

2 10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.475975089062 | 0.456449958437 | 0.520574925938 |
| 0.527724864062 | 0.586549870000 | 0.486725134688 |
| 0.597624945000 | 0.407399910000 | 0.549425000625 |
| 0.399974865312 | 0.571799980313 | 0.483674886250 |
| 0.641625000000 | 0.470749834687 | 0.615774896875 |
| 0.592525028750 | 0.563750015312 | 0.608125022500 |
| 0.629225023125 | 0.425949952500 | 0.451225058125 |
| 0.480674914688 | 0.390149985000 | 0.412774932813 |
| 0.358375000000 | 0.481049866875 | 0.471875045313 |

|                |                |                |
|----------------|----------------|----------------|
| 0.493124814375 | 0.550799825312 | 0.600824961875 |
| 0.394375012813 | 0.435250112188 | 0.384225103125 |
| 0.554625116563 | 0.481749848437 | 0.409374870625 |
| 0.626074929063 | 0.609850015000 | 0.405125058438 |

#### 10<sup>th</sup> delithiation

S<sub>10</sub>I

1.0

|                  |                  |                  |
|------------------|------------------|------------------|
| 20.0000000000000 | 0.0000000000000  | 0.0000000000000  |
| 0.0000000000000  | 20.0000000000000 | 0.0000000000000  |
| 0.0000000000000  | 0.0000000000000  | 20.0000000000000 |

S I

10 1

Direct

|                |                |                |
|----------------|----------------|----------------|
| 0.579200072000 | 0.446724939875 | 0.567724921250 |
| 0.345650000000 | 0.501974908250 | 0.549175138250 |
| 0.564649883375 | 0.537074870000 | 0.613975127000 |
| 0.490050213125 | 0.589725198500 | 0.565525047875 |
| 0.654350000000 | 0.461674895000 | 0.496524809750 |
| 0.422050163000 | 0.496875184250 | 0.407624997875 |
| 0.383150104250 | 0.410274801500 | 0.547824961625 |
| 0.402649911500 | 0.564225035000 | 0.612325125500 |
| 0.439250155250 | 0.412274791625 | 0.448175058125 |
| 0.613099962500 | 0.470925090500 | 0.405924832625 |
| 0.585149878625 | 0.588224916500 | 0.386024873000 |

Li<sub>20</sub>S<sub>10</sub>I supported over graphene

1.0

|               |               |              |
|---------------|---------------|--------------|
| 17.2576007843 | 0.0000000000  | 0.0000000000 |
| -8.6238807673 | 14.9436254716 | 0.0000000000 |

0.0000000000 0.0000000000 16.7147998810

C Li S I

98 20 10 1

Direct

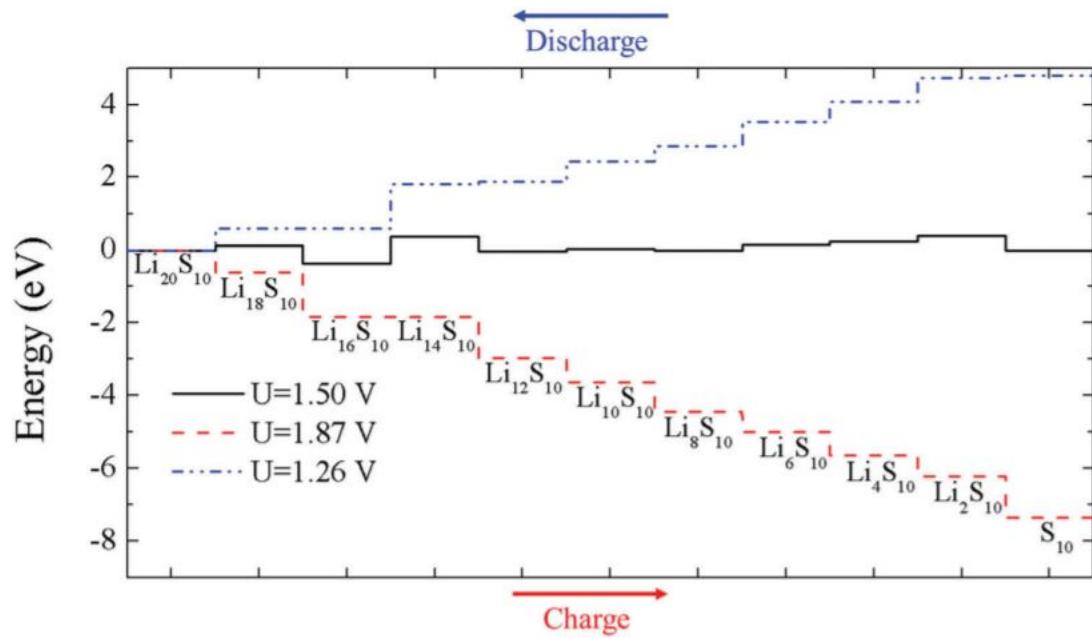
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|-------------|-------------|-------------|
| 0.059564197 | 0.047622999 | 0.046366999 |
| 0.107159006 | 0.000007028 | 0.046614400 |
| 0.202329985 | 0.047562001 | 0.044141100 |
| 0.249989003 | 0.000000000 | 0.045256799 |
| 0.345214978 | 0.047526602 | 0.043493900 |
| 0.392863039 | 0.999973705 | 0.045130499 |
| 0.488096005 | 0.047593300 | 0.044455901 |
| 0.535705989 | 0.999978747 | 0.045747999 |
| 0.630926009 | 0.047617000 | 0.045897599 |
| 0.678522040 | 0.999976960 | 0.046621600 |
| 0.773751986 | 0.047590400 | 0.047251601 |
| 0.821394037 | 0.999992468 | 0.047716901 |
| 0.916642016 | 0.047570002 | 0.047781699 |
| 0.964324960 | 0.000026300 | 0.047954399 |
| 0.059481103 | 0.190308010 | 0.045474998 |
| 0.107142002 | 0.142789999 | 0.044047700 |
| 0.202236999 | 0.190303989 | 0.040272197 |
| 0.249786992 | 0.142680998 | 0.040215498 |
| 0.345073996 | 0.190248005 | 0.037203700 |
| 0.392819002 | 0.142652998 | 0.039570302 |
| 0.488079013 | 0.190292996 | 0.038470700 |
| 0.535749950 | 0.142792999 | 0.041801899 |
| 0.631012995 | 0.190488998 | 0.042225802 |
| 0.678569014 | 0.142847005 | 0.044946801 |
| 0.773780967 | 0.190460998 | 0.045884501 |
| 0.821389986 | 0.142844005 | 0.047135800 |

|             |             |             |
|-------------|-------------|-------------|
| 0.916633997 | 0.190415990 | 0.047316702 |
| 0.964277006 | 0.142800003 | 0.047178998 |
| 0.059467805 | 0.333272982 | 0.046513301 |
| 0.107043993 | 0.285556992 | 0.044842600 |
| 0.202350011 | 0.333191008 | 0.041417497 |
| 0.249916001 | 0.285483984 | 0.037886200 |
| 0.345075027 | 0.333256996 | 0.033570701 |
| 0.392795005 | 0.285632999 | 0.032889602 |
| 0.488167989 | 0.333374995 | 0.030988000 |
| 0.535797969 | 0.285639987 | 0.034455998 |
| 0.631183027 | 0.333286990 | 0.036020101 |
| 0.678699023 | 0.285662994 | 0.040204700 |
| 0.773832061 | 0.333274992 | 0.042733401 |
| 0.821384998 | 0.285686989 | 0.045294898 |
| 0.916593993 | 0.333262995 | 0.046718601 |
| 0.964209054 | 0.285653006 | 0.047171499 |
| 0.059497615 | 0.476154002 | 0.047724197 |
| 0.107153006 | 0.428562999 | 0.046570000 |
| 0.202408020 | 0.476248995 | 0.044666701 |
| 0.250011011 | 0.428583006 | 0.041387101 |
| 0.345331023 | 0.476244017 | 0.037327800 |
| 0.392849011 | 0.428541014 | 0.032689401 |
| 0.488128986 | 0.476220022 | 0.030701000 |
| 0.535785992 | 0.428607991 | 0.029913601 |
| 0.630993024 | 0.476030003 | 0.033658003 |
| 0.678749961 | 0.428528984 | 0.035847900 |
| 0.773792070 | 0.475962005 | 0.040689599 |
| 0.821384046 | 0.428429013 | 0.043113702 |
| 0.916564050 | 0.476042990 | 0.046360798 |
| 0.964251076 | 0.428486003 | 0.047207098 |

|             |             |             |
|-------------|-------------|-------------|
| 0.059557605 | 0.618996029 | 0.049142099 |
| 0.107197017 | 0.571386998 | 0.048294900 |
| 0.202435017 | 0.619044020 | 0.047415301 |
| 0.250089998 | 0.571511954 | 0.045114599 |
| 0.345281988 | 0.619140960 | 0.042842499 |
| 0.392936019 | 0.571615977 | 0.038588099 |
| 0.488171030 | 0.619232986 | 0.036952202 |
| 0.535560053 | 0.571400017 | 0.034167403 |
| 0.630762004 | 0.618907960 | 0.038209000 |
| 0.678471024 | 0.571243981 | 0.038542800 |
| 0.773715043 | 0.618889963 | 0.043716599 |
| 0.821355057 | 0.571167017 | 0.044275399 |
| 0.916670973 | 0.618930998 | 0.047959202 |
| 0.964292059 | 0.571273018 | 0.048133197 |
| 0.059577504 | 0.761912978 | 0.049869398 |
| 0.107174006 | 0.714250978 | 0.049595000 |
| 0.202448016 | 0.761900023 | 0.048841397 |
| 0.250072990 | 0.714292013 | 0.047778300 |
| 0.345293975 | 0.761902959 | 0.046485900 |
| 0.392920010 | 0.714326985 | 0.044174802 |
| 0.488099036 | 0.761888025 | 0.043092898 |
| 0.535645065 | 0.714366042 | 0.040365202 |
| 0.630752025 | 0.761991985 | 0.042261900 |
| 0.678402034 | 0.714283015 | 0.042401002 |
| 0.773787966 | 0.761897981 | 0.046126599 |
| 0.821408990 | 0.714196031 | 0.046824803 |
| 0.916681011 | 0.761884005 | 0.049013203 |
| 0.964301992 | 0.714253020 | 0.049479398 |
| 0.059559022 | 0.904786021 | 0.048720799 |
| 0.107176024 | 0.857167034 | 0.049085699 |

|             |             |             |
|-------------|-------------|-------------|
| 0.202429016 | 0.904793041 | 0.047811100 |
| 0.250038006 | 0.857155036 | 0.048066799 |
| 0.345263994 | 0.904758962 | 0.046928598 |
| 0.392872028 | 0.857136018 | 0.046660800 |
| 0.488096008 | 0.904738029 | 0.046114603 |
| 0.535681037 | 0.857102003 | 0.045146500 |
| 0.630877978 | 0.904748049 | 0.045729498 |
| 0.678462033 | 0.857125999 | 0.045181200 |
| 0.773720994 | 0.904752006 | 0.047055501 |
| 0.821427026 | 0.857220003 | 0.047711499 |
| 0.916689982 | 0.904789977 | 0.048690000 |
| 0.964311087 | 0.857164992 | 0.049233199 |
| 0.371437035 | 0.457499004 | 0.431264021 |
| 0.644329029 | 0.559977017 | 0.437837007 |
| 0.619839047 | 0.307098006 | 0.355075999 |
| 0.499278999 | 0.369578992 | 0.352542013 |
| 0.370940027 | 0.260147003 | 0.201050004 |
| 0.804364974 | 0.567103014 | 0.234472011 |
| 0.473381001 | 0.262782996 | 0.459628997 |
| 0.317243003 | 0.197924002 | 0.350587000 |
| 0.428677023 | 0.604764010 | 0.347628007 |
| 0.715128993 | 0.484342986 | 0.361635006 |
| 0.494910998 | 0.434916988 | 0.499550972 |
| 0.543967036 | 0.544742990 | 0.310283979 |
| 0.563917005 | 0.325010001 | 0.207675995 |
| 0.637692976 | 0.492460014 | 0.226228991 |
| 0.419991001 | 0.520306994 | 0.209896003 |
| 0.615812010 | 0.693967979 | 0.364581989 |
| 0.219977012 | 0.346545016 | 0.220836002 |
| 0.665431954 | 0.679240987 | 0.209432012 |

|             |             |             |
|-------------|-------------|-------------|
| 0.229923999 | 0.296842017 | 0.376154992 |
| 0.360026996 | 0.366133993 | 0.301207989 |
| 0.608703979 | 0.408466988 | 0.455915984 |
| 0.713922027 | 0.413252997 | 0.244804995 |
| 0.459310013 | 0.224418995 | 0.297965001 |
| 0.233437005 | 0.225990995 | 0.255807013 |
| 0.361889984 | 0.316585976 | 0.444810980 |
| 0.518145042 | 0.575839972 | 0.448687999 |
| 0.719615994 | 0.621383019 | 0.307389002 |
| 0.485655974 | 0.418018999 | 0.221971011 |
| 0.533981007 | 0.666331982 | 0.245692011 |
| 0.299904003 | 0.461430999 | 0.311358993 |
| 0.518280053 | 0.133618998 | 0.424616007 |



**Figure S1.** Energy profiles of calculated delithiation (charge) at different applied voltages for the  $\text{Li}_{20}\text{S}_{10}$  cluster. The arrows indicate the directions of charge and discharge. Figure 1 reproduced from reference 20 with permission from the PCCP Owner Societies.