# Peri-diselenolo-substituted 1,8-naphthalimide derivatives as bipolar matrices for redox reactions in a non-aqueous electrolyte

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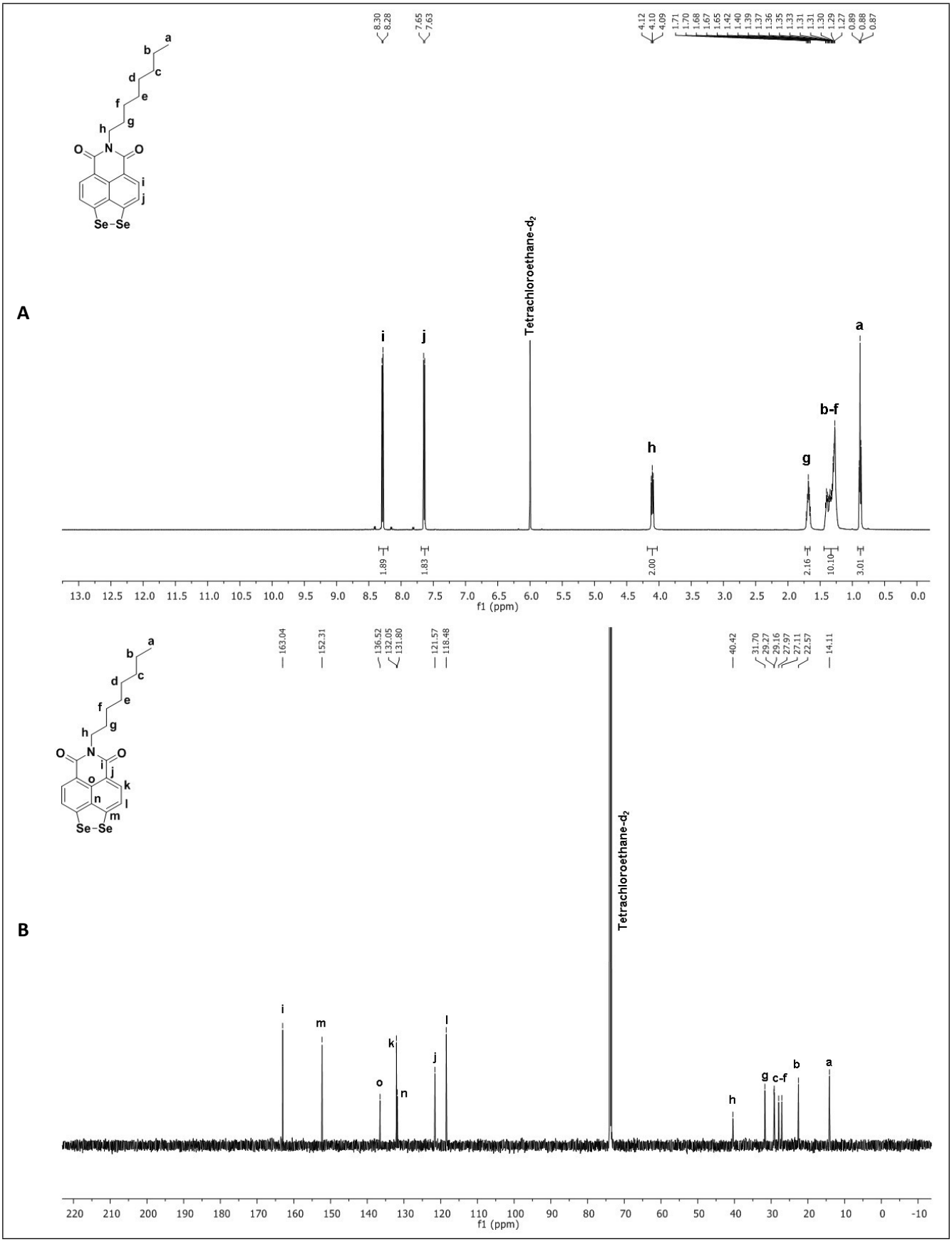

A
B
Tetrachloroethane-d2
f1 (ppm)

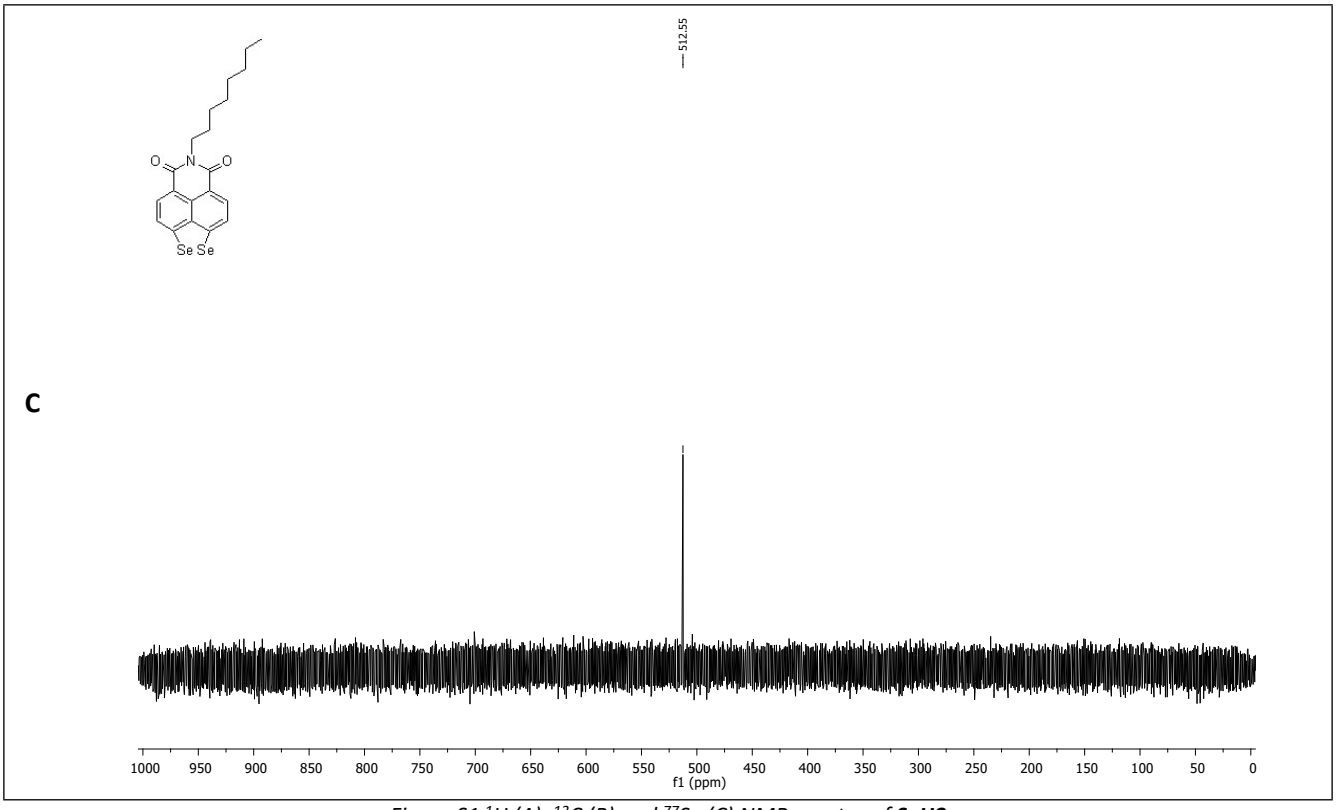
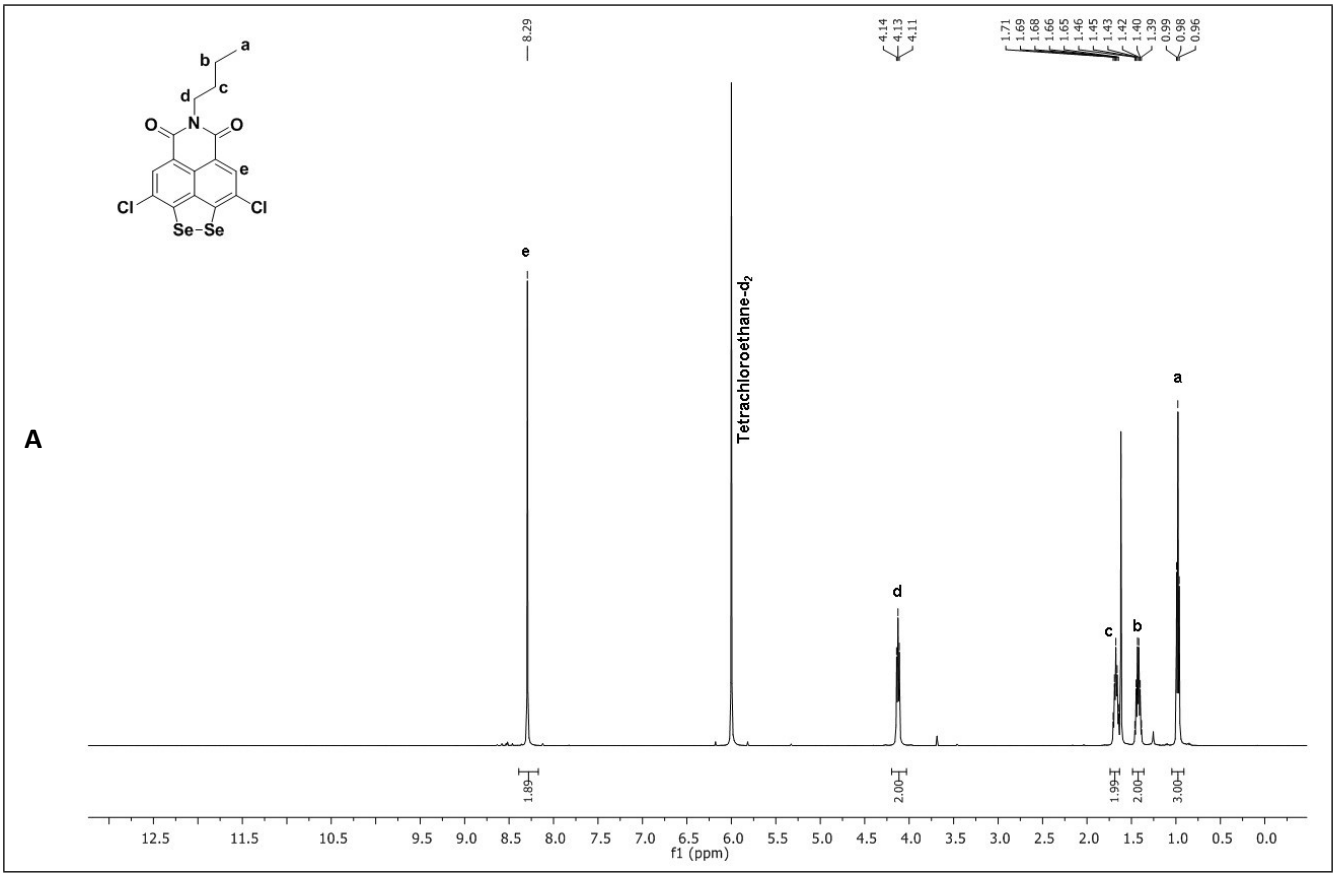



*Figure S1 $^{1}H$ (A), $^{13}C$ (B) and $^{77}Se$ (C) NMR spectra of* ***SeH8***



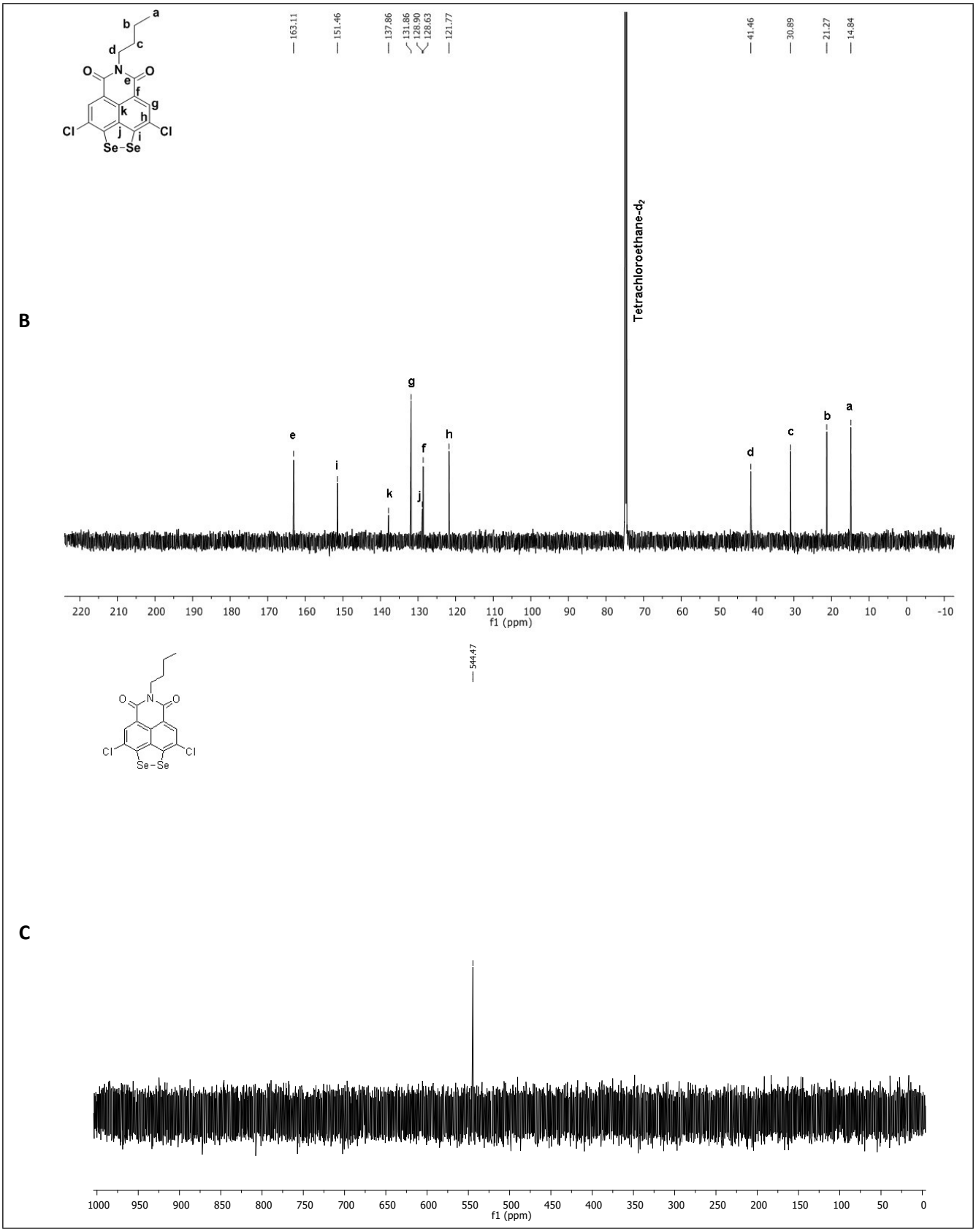
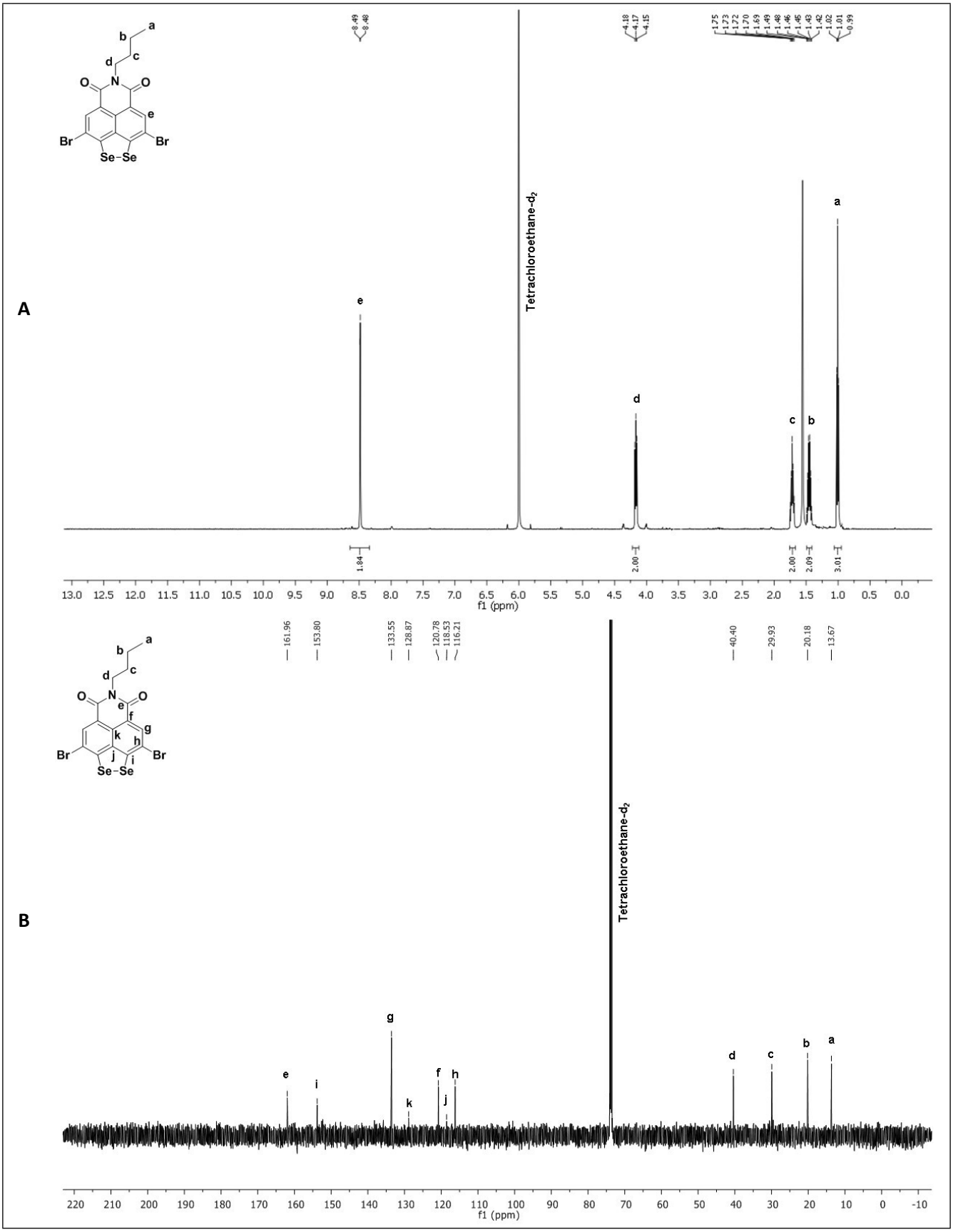



*Figure S2 $^{1}H$ (A), $^{13}C$ (B) and $^{77}Se$ (C) NMR spectra of **SeCl4***


A
B
Tetrachloroethane-d2
f1 (ppm)
Br
Se–Se
O
N

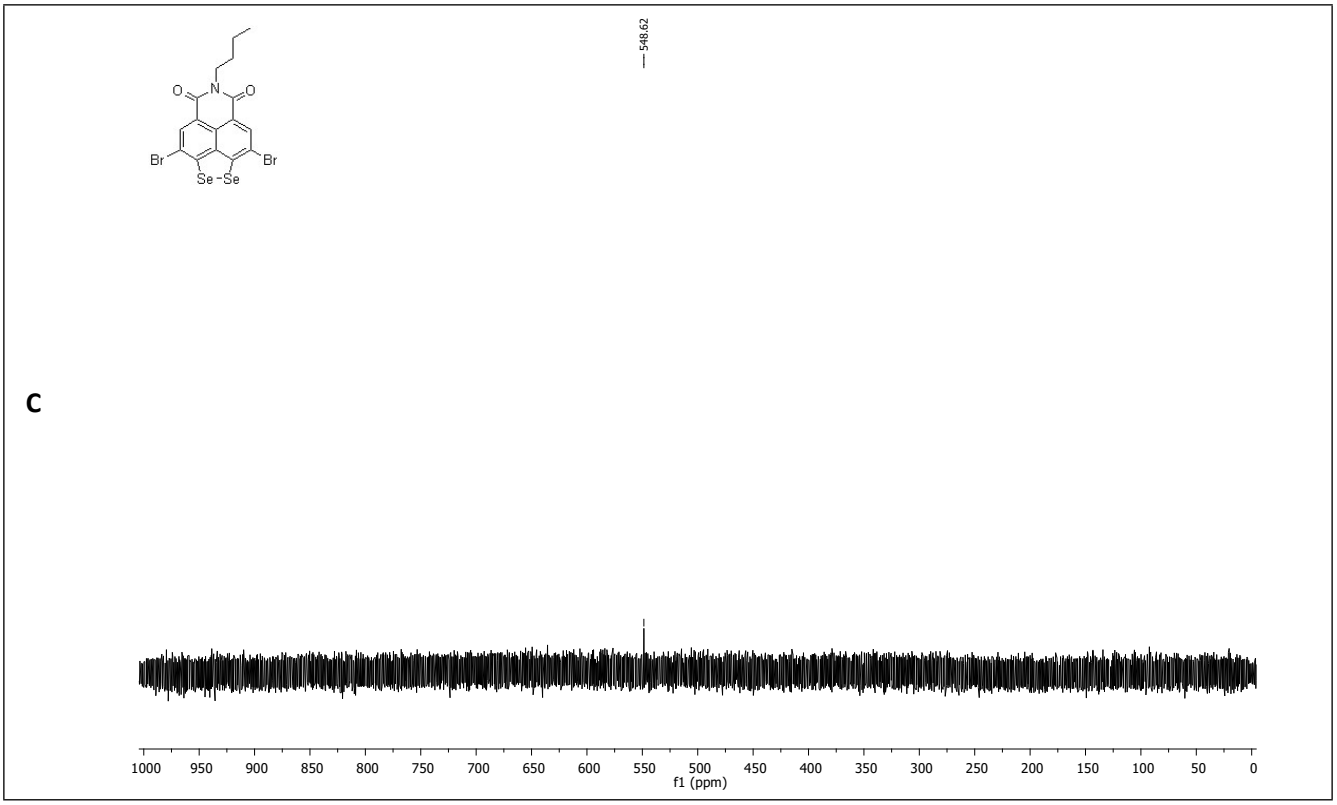



*Figure S3 $^{1}H$ (A), $^{13}C$ (B) and $^{77}Se$ (C) NMR spectra of **SeBr4***

*Table S1. Dipole moments (in Debye) of the neutral models*

| derivative | Dipole moment, D |
|---|---|
| | neutral |
| **SeH** | 7.1 |
| **SeCl** | 5.1 |
| **SeBr** | 5.2 |

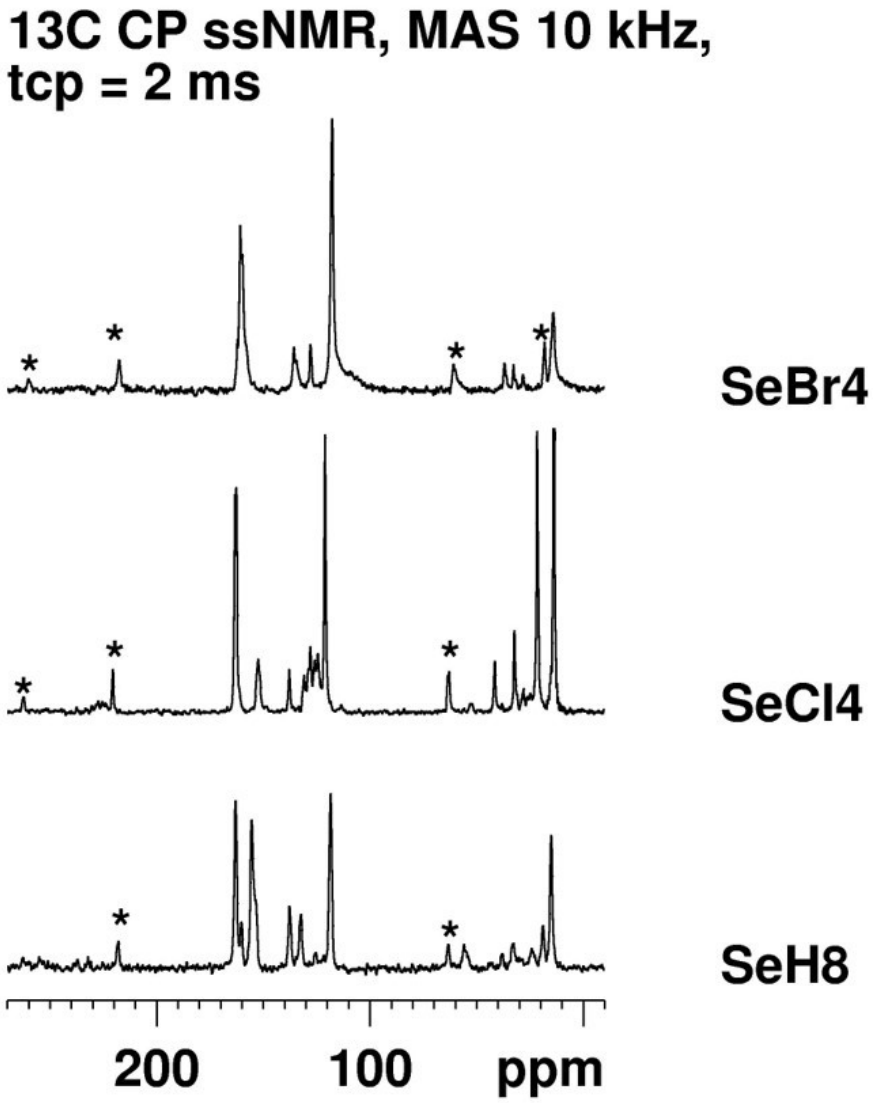



*Figure S4 $^{13}C$ cross-polarization (CP) solid state NMR spectra of the **SeBr4**, **SeCl4** and **SeH8** derivatives recorded with 2 ms contact time*

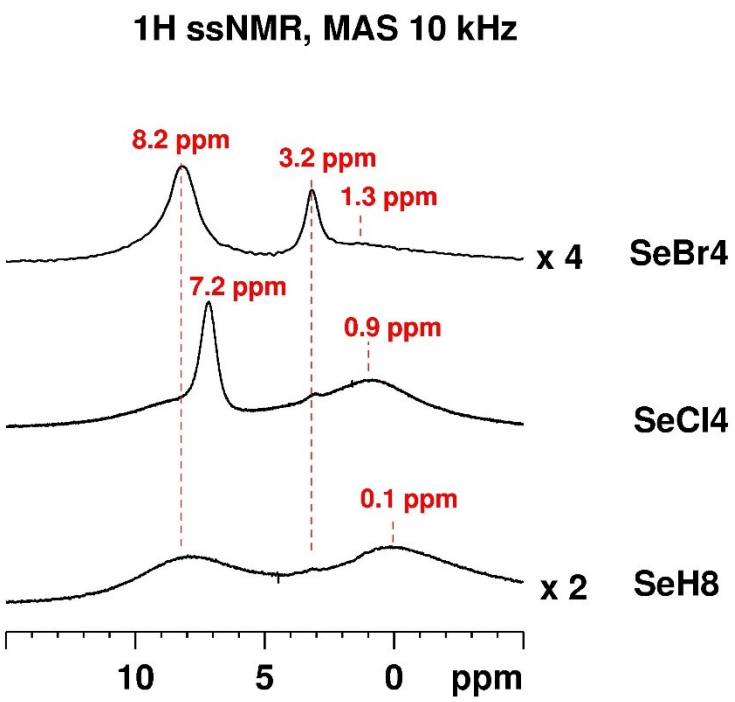



*Figure S5 $^{1}H$ solid state NMR spectra of the **SeBr4**, **SeCl4** and **SeH8** derivatives recorded with background suppression*

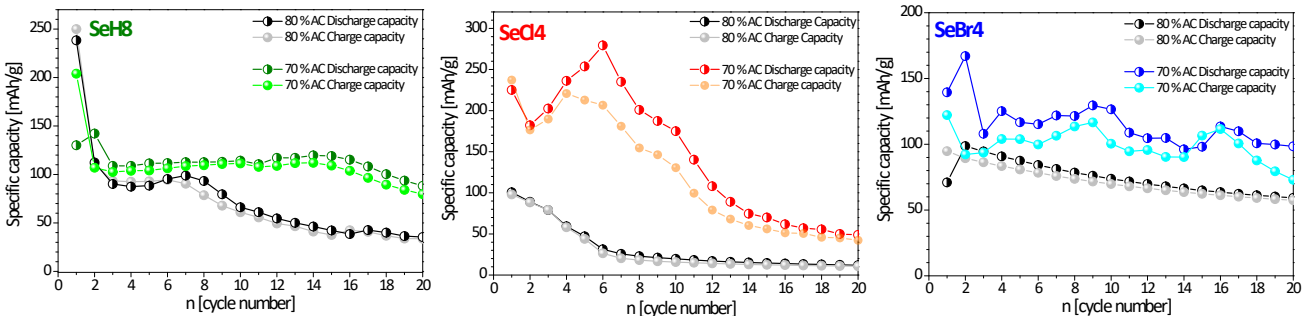
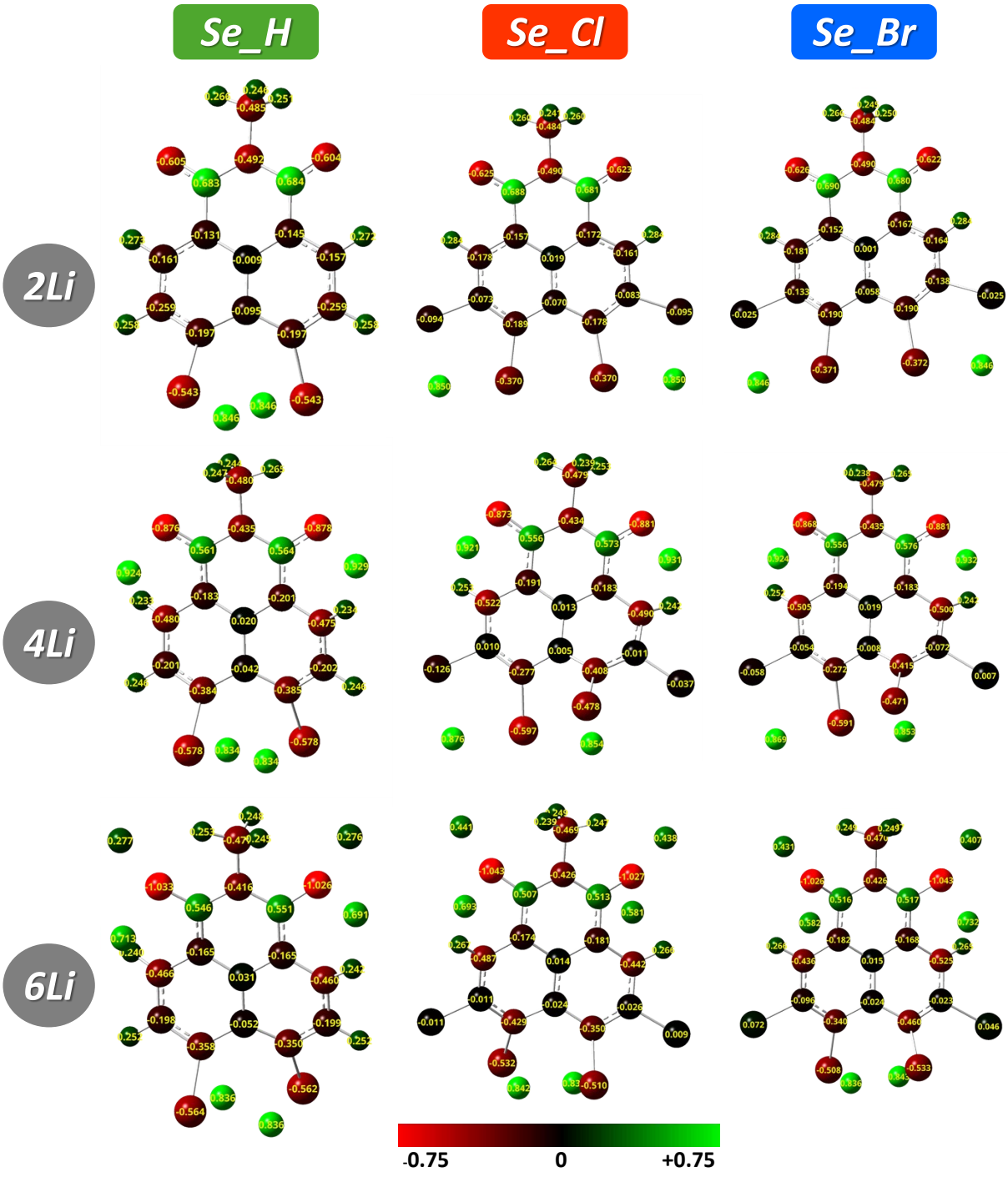



*Figure S6 Cycling stability of **SeH8**, **SeCl4** and **SeBr4** (80% and 70% active compound) used as electrodes in lithium half-cell starting with a discharge mode at scan rate C/10.*

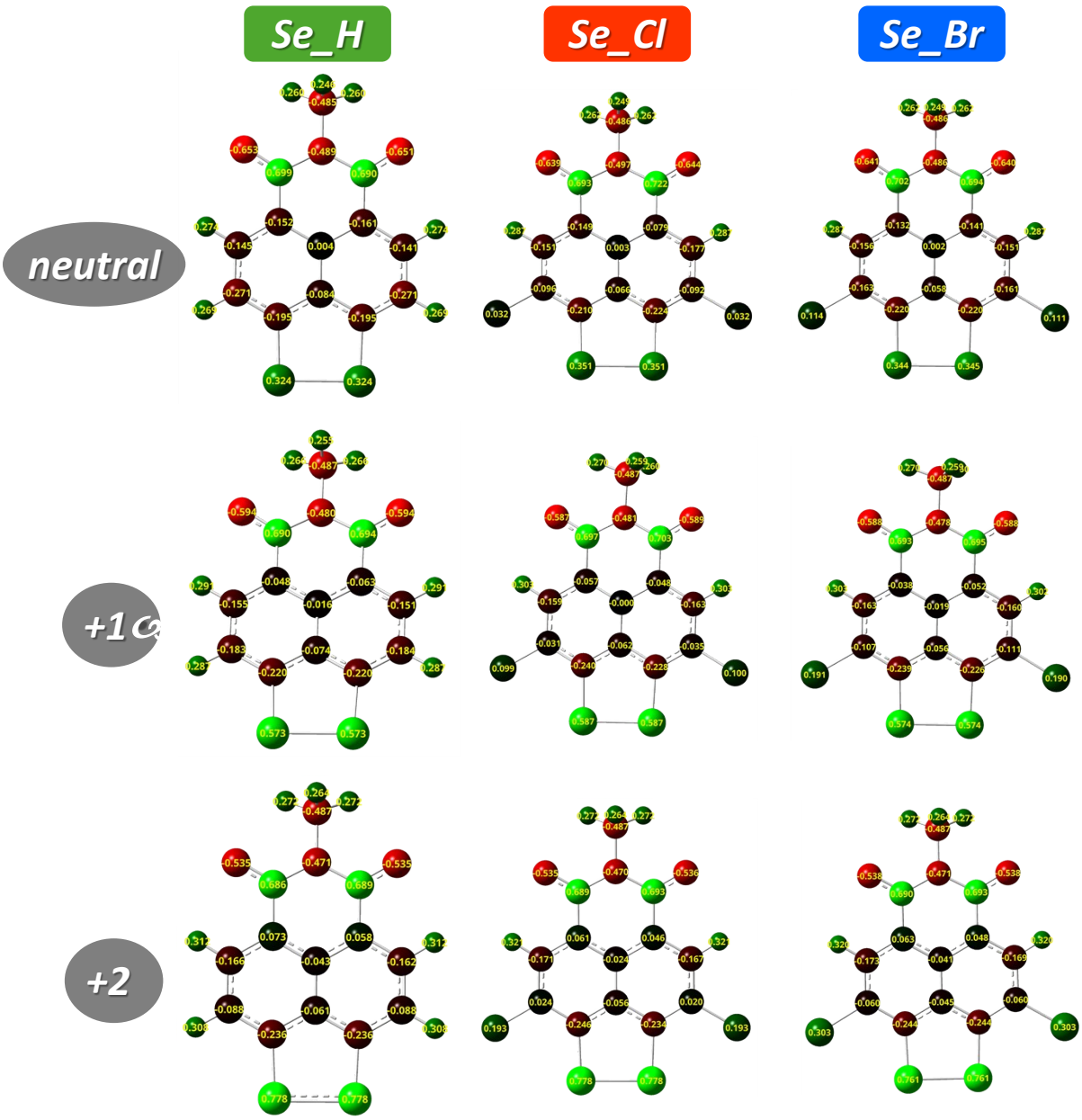
*NBO charges reduction*

**A**



*NBO charges without TFSI*

**B**



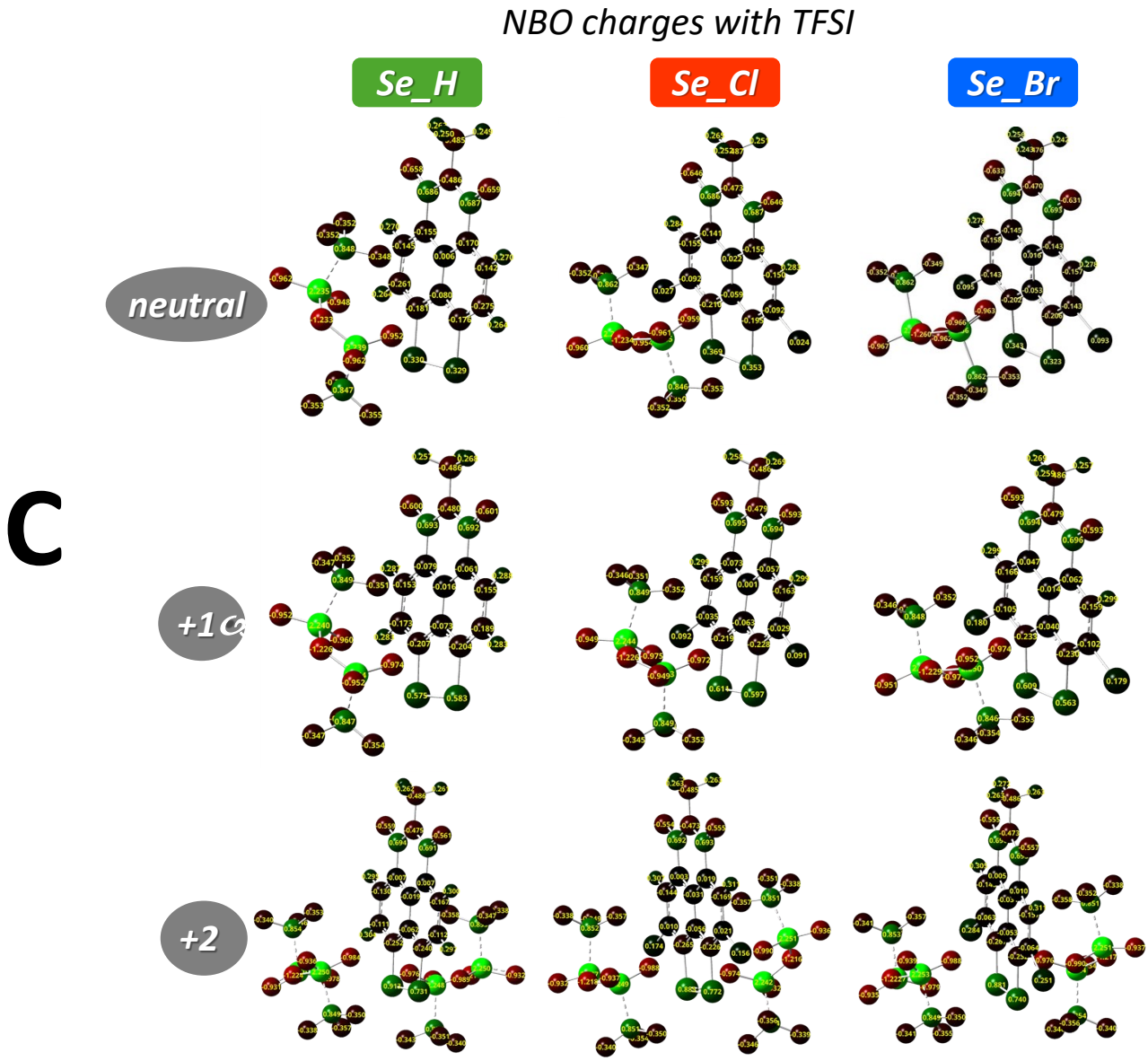



*Figure S7 NBO charges of SeH, SeCl, and SeBr after the reduction with 2Li, 4Li and 6Li (A). NBO charges of SeH, SeCl, and SeBr after the oxidation without $TFSI^-$ (B), and with $TFSI^-$ (C)*

*Table S2a. Se-Se distance in Å as a function of the number of inserted $Li^+$*

| Se-Se distance in Å | 0Li | 2Li | 4Li |
|---|---|---|---|
| **SeH** | 2.37 | 3.68 | 3.76 |
| **SeCl** | 2.39 | 3.14 | 3.66 |
| **SeBr** | 2.39 | 3.13 | 3.66 |

*Table S2b The electron distribution of the imide chain*

| Summed charges on the imide chain (O=C-N-C=O) | 0Li | 2Li | 4Li | 6Li |
|---|---|---|---|---|
| **SeH** | -0.40 | -0.33 | -1.05 | -1.38 |
| **SeCl** | -0.36 | -0.37 | -1.06 | -1.48 |
| **SeBr** | -0.37 | -0.37 | -1.05 | -1.46 |

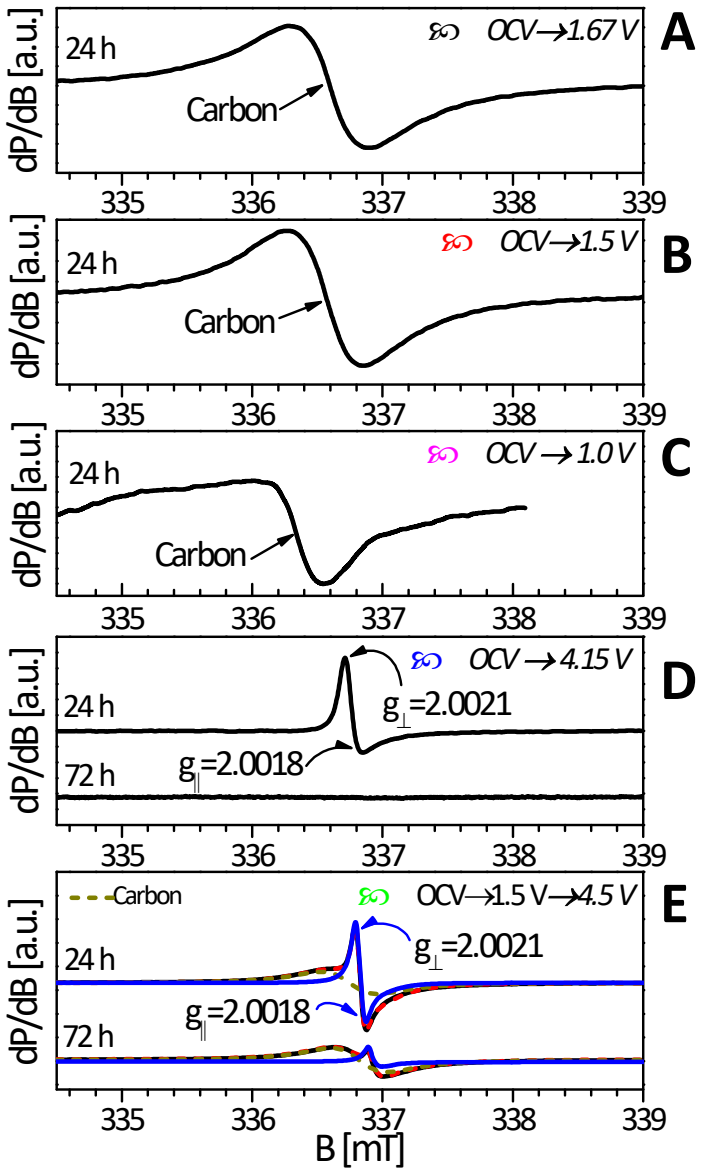



*Figure S8 EPR spectra of **SeCl4** electrodes discharged consecutively to 1.67 V (A), to 1.50 V (B) and to 1.00 V (C). The EPR spectra of **SeCL4** charged directly to 4.15 V (D) and discharged to 1.5 V first, followed by charge to 4.5 V (E). The EPR spectra are detected after 24h and 72h of the electrochemical experiments.*

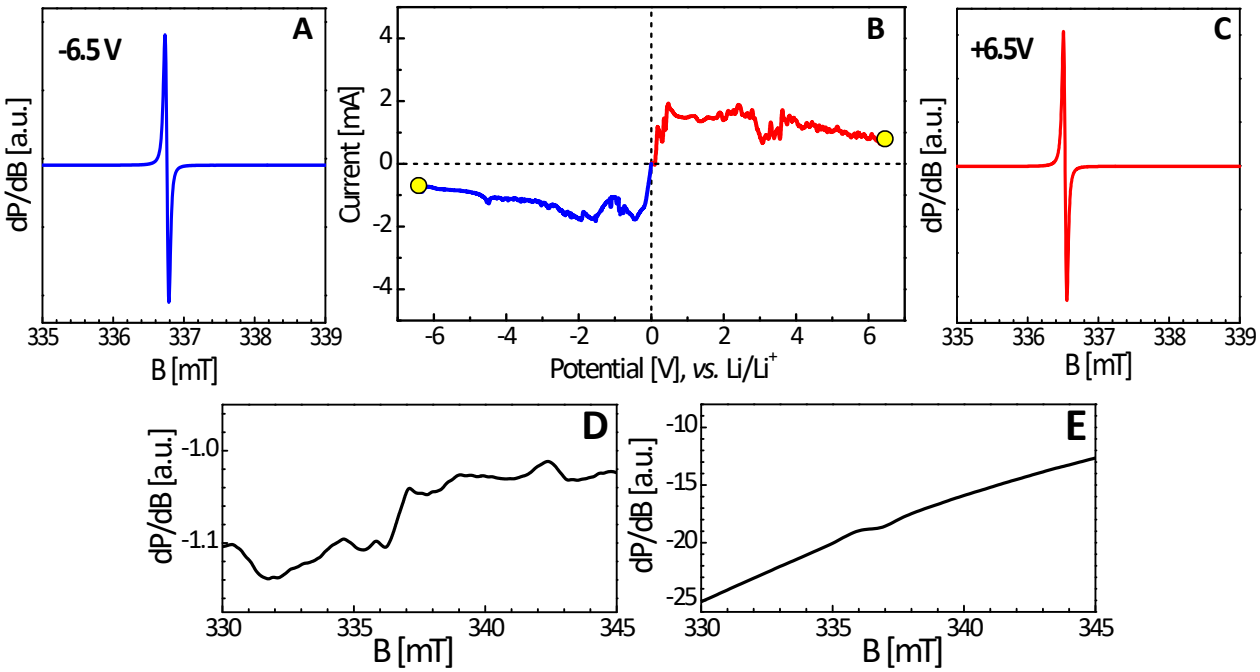



*Figure S9 Linear sweep voltammetry curves of the symmetrical cell Li|Li|LiTFSI-Pyr13FSI|Li (B). Ex-situ EPR spectra of the separator soaked in the electrolyte solution after the cathodic (A) and anodic (C) scans at the given potential. For the sake of comparison, the EPR spectra of blank electrolyte (D) and the unsoaked separator (E) are also given.*

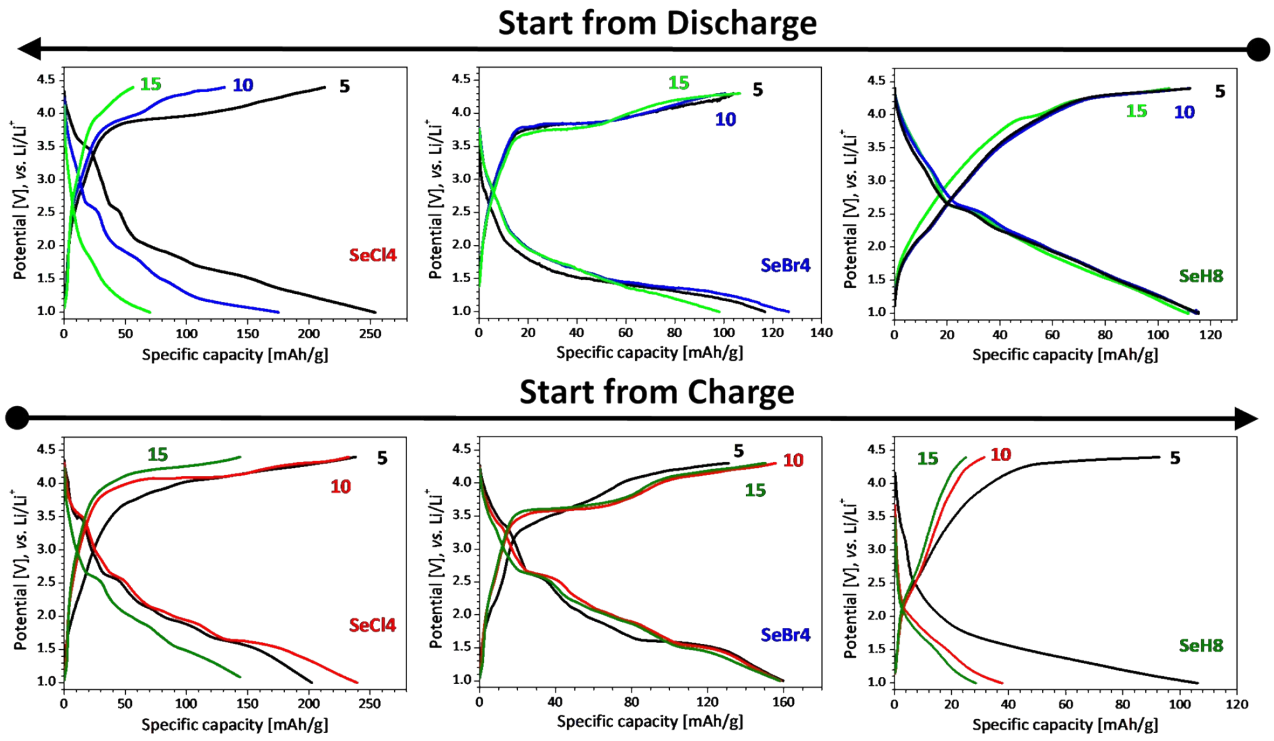



*Figure S10 Charge/discharge curves of **SeCl4**, **SeBr4** and **SeH8** used as electrodes in lithium half-cell starting with a discharge and charge mode at scan rate C/10 (5th, 10th and 15th cycle).*

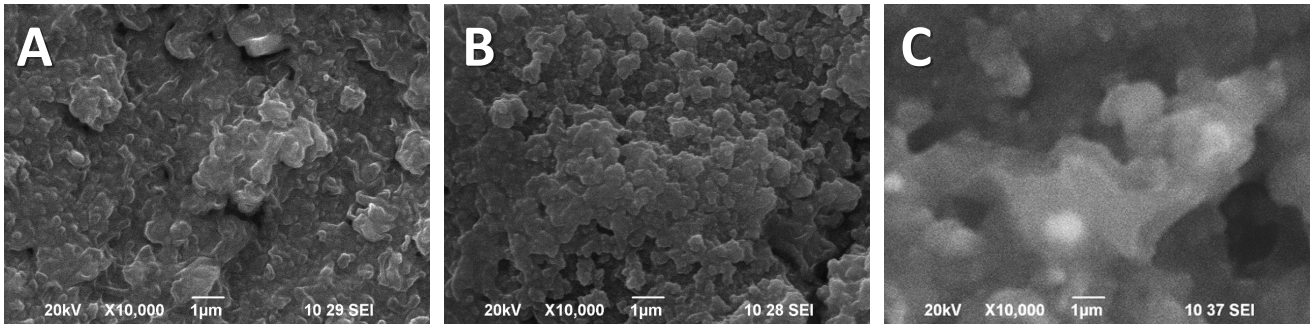



*Figure S11 SEM images of **SeCl4** (A), **SeBr4** (B) and **SeH8** (C) electrodes cycled for 50 times in LiTFSI:$Pyr_{1,3}$FSI electrolyte.*

*Table S3 EDS data in at% (averaged values) for the pristine compounds SeCl4, SeBr4 and SeH8*

| Element, at % | SeCl4 | SeBr4 | SeH8 |
|---|---|---|---|
| C | 78.27 | 76.46 | 82.10 |
| N | 2.89 | 3.59 | 2.99 |
| O | 8.98 | 7.55 | 8.80 |
| Cl | 4.88 | - | - |
| Se | 4.99 | 6.36 | 6.12 |
| Br | - | 6.94 | - |

*Table S4 EDS data in at% (averaged values) for the electrodes cycled for 50 times in LiTFSI:$Pyr_{1,3}$FSI electrolyte*

| Element, at % | SeCl4 electrode | SeBr4 electrode | SeH8 electrode |
|---|---|---|---|
| C | 38.06 | 44.27 | 47.95 |
| N | 2.88 | 6.31 | 6.46 |
| O | 39.19 | 27.95 | 22.63 |
| F | 10.73 | 12.29 | 12.89 |
| Na | 2.60 | 0.79 | 0.78 |
| S | 4.53 | 7.57 | 7.70 |
| Cl | 0.65 | - | - |
| Se | 1.34 | 0.29 | 1.01 |
| Br | - | 0.53 | - |