# Supporting Information

**Evaluation of Computational Chemistry Methods for Predicting Redox Potentials of Quinone-based Cathodes for Li-ion Batteries**

Xuan Zhou [1,2,3], Abhishek Khetan [1,2,4], and Süleyman Er [1,2,*]

[1] DIFFER – Dutch Institute for Fundamental Energy Research, De Zaale 20, 5612 AJ Eindhoven, the Netherlands

[2] CCER – Center for Computational Energy Research, De Zaale 20, 5612 AJ Eindhoven, the Netherlands

[3] Department of Applied Physics, Eindhoven University of Technology, 5600 MB Eindhoven, the Netherlands

[4] Present address: Fuel Science Center, RWTH Aachen University, 52062 Aachen, Germany

[*] E-mail: S.Er@differ.nl

The dielectric constant, molecular weight and density of binary solvents can be calculated by the below three equations using the available information on pure solvents [S1].

For a mixture solvent the dielectric constant, $\varepsilon_{mix}$, is calculated as [S2,S3]:

$$\varepsilon_{mix} = \sum x_i \, \varepsilon_i$$

where $x_i$ is the molar ratio and $\varepsilon_i$ is the dielectric constant of solvent $i$, respectively,

The molecular weight is determined as the equation:

$$M_{mix} = \sum x_i \, M_i$$

where $M_{mix}$ and $M_i$ represent the molecular density of a binary solvent and solvent i, separately.

The density of the mixture liquids can be given by:

$$\rho_{mix} = \left( \sum \frac{w_i}{\rho_i} \right)^{-1}$$

where $\rho_{mix}$ and $\rho_i$ are the density of mixture liquids, and solvent i. $w_i$ is the weight fraction of pure solvent.

**Table S1.** Physical properties, including dielectric constant, molecular weight, and density, of organic electrolytes and their mixtures corresponding to the experimental datasets.

| Solvent | $\varepsilon$ (25 °C) | Molecular weight (g/mol) | Density (g/cm³) |
| --- | --- | --- | --- |
| Ethylene carbonate | 89.78 | 88.06 | 1.32 |
| Diethyl carbonate | 2.81 | 118.13 | 0.97 |
| Dimethyl carbonate | 3.11 | 90.08 | 1.06 |
| Tetraglyme | 7.79 | 222.28 | 1.01 |
| Propylene carbonate | 64.92 | 102.09 | 1.20 |
| EC+DEC (v/v=3:7) | 41.00 | 104.93 | 1.08 |
| EC+DMC (w/w=1:1) | 46.98 | 89.06 | 1.18 |
| EC+DMC (v/v=1:1) | 51.66 | 88.95 | 1.19 |
| EC+DMC (v/v=3:7) | 33.71 | 89.37 | 1.14 |

The abbreviations of organic solvents in the above table are denoted as: EC = ethylene carbonate, DEC = diethyl carbonate, DMC = dimethyl carbonate, PC = propylene carbonate. w/w and v/v indicate the mass fraction and volume fraction of two solvents, respectively.
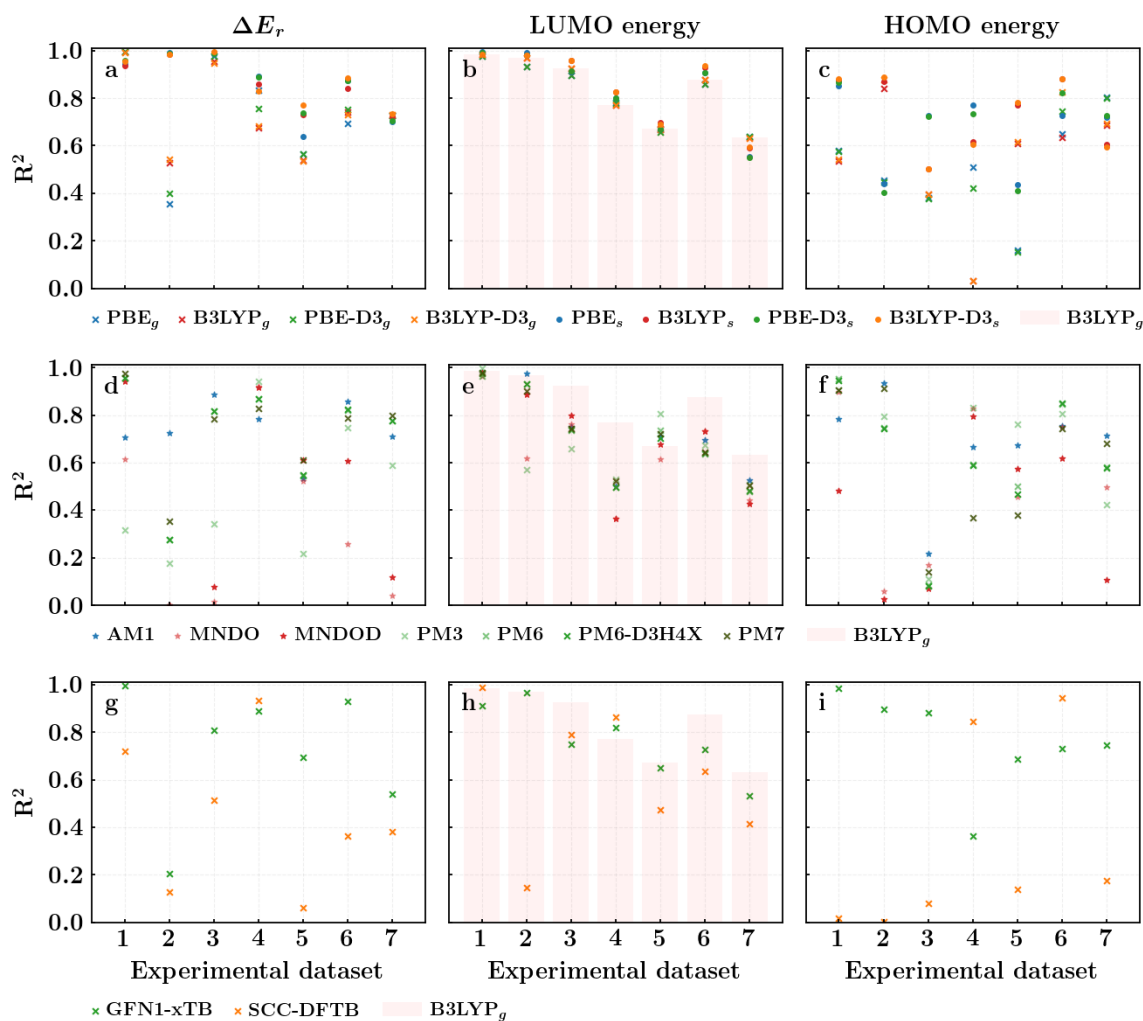
**Figure S1.** $R^2$ of three descriptors: $\Delta E_r$, LUMO energy, and HOMO energy. The computational data is calculated by using (a-c) DFT, (d-f) SEQM, and (g-i) DFTB methods for the seven experimental datasets. PBE$_g$ and PBE$_s$ represent single point calculations in gas ($g$) and solvent ($s$) phases, respectively. The shaded vertical bars in (b, e and h) show the fully B3LYP$_g$ calculated data.
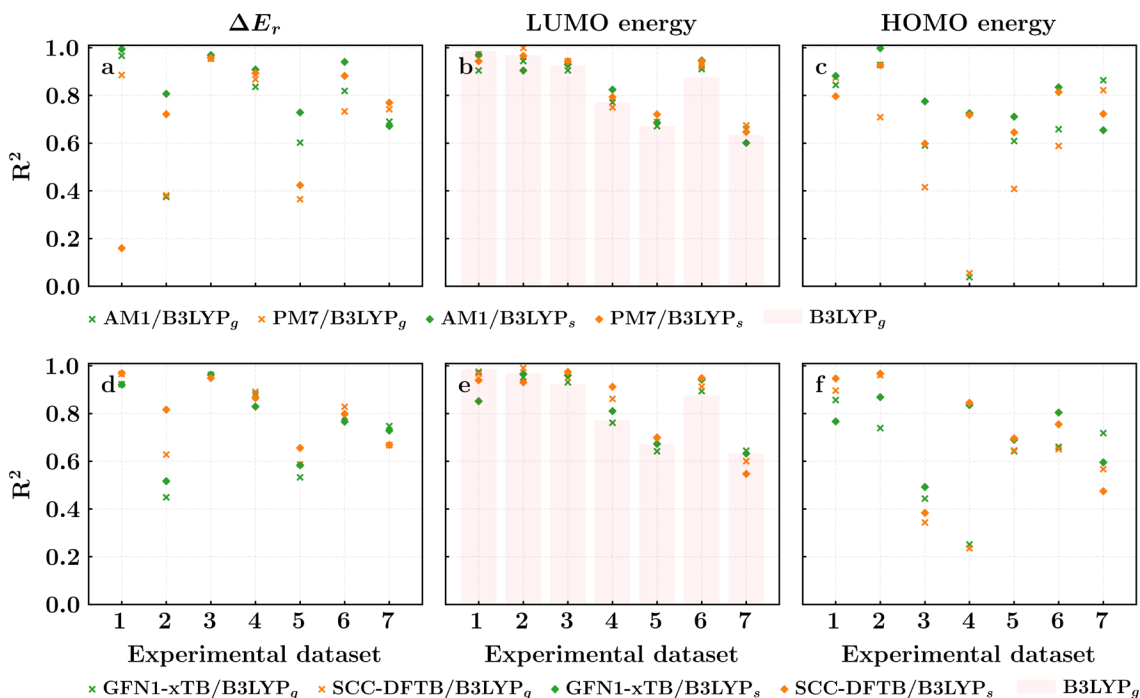
**Figure S2.** $R^2$ of three descriptors: $\Delta E_r$, LUMO energy, and HOMO energy. The computational data is calculated by using (a-c) hybrid SEQM/DFT scheme and (d-e) hybrid DFTB/DFT scheme for the seven experimental datasets. Accordingly, AM1/B3LYP$_g$ and AM1/B3LYP$_s$ imply that the molecules are optimized only with AM1 and their energies are calculated by using B3LYP functional either without or with solvation effect, respectively. The shaded vertical bars in (b, e and h) show the fully B3LYP$_g$ calculated data.

**Table S2.** NRMSE of $\Delta E_r$ as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 3.17% | 29.78% | 5.10% | 13.54% | 21.32% | 19.37% | 18.29% |
| | **B3LYP$_g$** | 3.12% | 25.47% | 7.13% | 18.90% | 21.96% | 17.72% | 18.24% |
| | **PBE-D3$_g$** | 3.24% | 28.79% | 5.13% | 16.45% | 21.34% | 17.36% | 18.22% |
| | **B3LYP-D3$_g$** | 3.87% | 25.09% | 7.47% | 18.67% | 22.04% | 18.13% | 18.12% |
| | **PBE$_s$** | 9.81% | 3.82% | 4.07% | 10.94% | 19.42% | 12.48% | 19.09% |
| | **B3LYP$_s$** | 10.65% | 4.86% | 2.94% | 12.45% | 16.81% | 13.93% | 18.32% |
| | **PBE-D3$_s$** | 8.39% | 4.02% | 3.90% | 11.10% | 16.50% | 12.34% | 18.96% |
| | **B3LYP-D3$_s$** | 8.72% | 4.54% | 2.70% | 13.76% | 15.52% | 11.89% | 18.01% |
| **SEQM** | **AM1** | 22.53% | 19.48% | 11.05% | 15.44% | 22.02% | 13.17% | 18.76% |
| | **MNDO** | 25.72% | 37.08% | 32.48% | 9.33% | 22.31% | 30.06% | 34.13% |
| | **MNDOD** | 9.94% | 37.07% | 31.46% | 9.60% | 20.14% | 21.89% | 32.74% |
| | **PM3** | 34.25% | 33.60% | 26.54% | 8.02% | 28.55% | 17.55% | 22.38% |
| | **PM6** | 8.48% | 31.58% | 14.01% | 12.12% | 21.74% | 14.71% | 16.53% |
| | **PM6-D3H4X** | 8.62% | 31.58% | 14.01% | 12.10% | 21.69% | 14.68% | 16.53% |
| | **PM7** | 6.66% | 29.81% | 15.22% | 13.72% | 20.10% | 16.11% | 15.61% |
| **DFTB** | **GFN1-xTB** | 2.70% | 33.04% | 14.33% | 11.02% | 17.81% | 9.27% | 23.66% |
| | **SCC-DFTB** | 21.88% | 34.60% | 22.82% | 8.56% | 31.26% | 27.86% | 27.42% |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 7.55% | 29.33% | 6.94% | 13.43% | 20.35% | 14.85% | 19.40% |
| | **PM7/B3LYP$_g$** | 14.03% | 29.17% | 7.09% | 12.06% | 25.72% | 18.02% | 17.71% |
| | **AM1/B3LYP$_s$** | 3.29% | 16.30% | 5.76% | 10.04% | 16.82% | 8.52% | 19.97% |
| | **PM7/B3LYP$_s$** | 38.01% | 19.57% | 6.79% | 10.83% | 24.50% | 11.99% | 16.74% |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 11.52% | 27.53% | 6.40% | 11.46% | 22.06% | 16.17% | 17.51% |
| | **SCC-DFTB/B3LYP$_g$** | 7.67% | 22.61% | 6.87% | 10.93% | 20.74% | 14.43% | 20.10% |
| | **GFN1-xTB/B3LYP$_s$** | 40.50% | 28.39% | 8.42% | 9.25% | 20.21% | 9.12% | 17.62% |
| | **SCC-DFTB/B3LYP$_s$** | 33.16% | 27.47% | 16.83% | 10.12% | 14.03% | 11.66% | 26.67% |

**Table S3.** NRMSE of LUMO energy of the reactant molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 6.40% | 9.76% | 10.59% | 15.49% | 18.64% | 13.09% | 21.02% |
| | **B3LYP$_g$** | 5.08% | 6.59% | 9.01% | 15.91% | 18.52% | 12.30% | 21.13% |
| | **PBE-D3$_g$** | 6.64% | 9.73% | 10.56% | 15.59% | 18.89% | 13.09% | 21.02% |
| | **B3LYP-D3$_g$** | 5.52% | 6.52% | 9.05% | 15.96% | 18.62% | 12.28% | 21.14% |
| | **PBE$_s$** | 2.90% | 3.53% | 9.82% | 14.81% | 18.57% | 10.73% | 23.29% |
| | **B3LYP$_s$** | 4.96% | 5.30% | 6.71% | 13.92% | 17.72% | 9.26% | 22.27% |
| | **PBE-D3$_s$** | 4.10% | 5.11% | 9.57% | 14.90% | 18.63% | 10.72% | 23.34% |
| | **B3LYP-D3$_s$** | 5.56% | 5.47% | 6.70% | 13.81% | 17.97% | 8.73% | 22.21% |
| **SEQM** | **AM1** | 6.21% | 6.08% | 16.35% | 23.45% | 17.36% | 19.29% | 23.97% |
| | **MNDO** | 7.76% | 22.95% | 16.04% | 26.45% | 20.02% | 18.00% | 26.07% |
| | **MNDOD** | 6.06% | 12.53% | 14.76% | 26.47% | 18.36% | 18.01% | 26.34% |
| | **PM3** | 2.57% | 24.29% | 19.11% | 22.70% | 14.19% | 19.94% | 24.40% |
| | **PM6** | 8.08% | 9.77% | 16.77% | 23.54% | 16.60% | 21.00% | 25.14% |
| | **PM6-D3H4X** | 6.50% | 9.77% | 16.73% | 23.57% | 17.57% | 20.88% | 25.12% |
| | **PM7** | 6.31% | 11.68% | 16.62% | 22.91% | 17.04% | 20.80% | 24.58% |
| **DFTB** | **GFN1-xTB** | 12.48% | 6.84% | 16.37% | 14.09% | 19.14% | 18.27% | 23.79% |
| | **SCC-DFTB** | 4.38% | 34.27% | 15.07% | 12.32% | 23.44% | 21.07% | 26.65% |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 12.81% | 8.78% | 10.10% | 15.88% | 18.50% | 10.43% | 20.33% |
| | **PM7/B3LYP$_g$** | 6.89% | 1.33% | 7.76% | 16.60% | 17.96% | 9.82% | 19.89% |
| | **AM1/B3LYP$_s$** | 7.13% | 11.49% | 8.63% | 13.90% | 18.06% | 8.09% | 22.02% |
| | **PM7/B3LYP$_s$** | 9.85% | 6.87% | 7.82% | 15.09% | 17.06% | 8.36% | 20.70% |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 6.57% | 8.13% | 8.59% | 16.20% | 19.32% | 11.36% | 20.78% |
| | **SCC-DFTB/B3LYP$_g$** | 7.54% | 3.56% | 7.28% | 12.35% | 18.12% | 10.30% | 22.03% |
| | **GFN1-xTB/B3LYP$_s$** | 15.95% | 6.85% | 6.51% | 14.44% | 18.46% | 8.41% | 21.12% |
| | **SCC-DFTB/B3LYP$_s$** | 10.18% | 9.75% | 5.22% | 9.85% | 17.69% | 7.90% | 23.45% |

**Table S4.** NRMSE of HOMO energy of the product molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 26.90% | 27.37% | 25.79% | 23.20% | 29.58% | 20.67% | 15.48% |
| | **B3LYP$_g$** | 28.25% | 14.86% | 25.49% | 32.66% | 20.18% | 21.07% | 19.54% |
| | **PBE-D3$_g$** | 26.98% | 27.60% | 25.86% | 25.26% | 29.69% | 17.58% | 15.55% |
| | **B3LYP-D3$_g$** | 28.05% | 13.09% | 25.44% | 32.67% | 20.04% | 14.60% | 19.29% |
| | **PBE$_s$** | 15.90% | 27.81% | 17.17% | 15.86% | 24.27% | 18.27% | 18.44% |
| | **B3LYP$_s$** | 14.78% | 13.30% | 23.07% | 20.60% | 15.52% | 12.11% | 21.87% |
| | **PBE-D3$_s$** | 15.26% | 28.67% | 17.29% | 17.08% | 24.77% | 14.71% | 18.22% |
| | **B3LYP-D3$_s$** | 14.37% | 12.50% | 23.07% | 20.82% | 15.05% | 12.09% | 22.23% |
| **SEQM** | **AM1** | 19.29% | 9.54% | 28.98% | 19.12% | 18.40% | 17.33% | 18.68% |
| | **MNDO** | 13.38% | 35.97% | 29.81% | 13.76% | 23.84% | 17.45% | 24.71% |
| | **MNDOD** | 29.80% | 36.62% | 31.58% | 14.99% | 21.09% | 21.57% | 32.92% |
| | **PM3** | 12.81% | 16.74% | 30.86% | 13.62% | 15.73% | 15.32% | 26.48% |
| | **PM6** | 8.97% | 18.64% | 31.33% | 21.20% | 22.83% | 13.63% | 22.55% |
| | **PM6-D3H4X** | 9.84% | 18.83% | 31.38% | 21.25% | 23.53% | 13.53% | 22.66% |
| | **PM7** | 12.81% | 10.88% | 30.36% | 26.40% | 25.41% | 17.68% | 19.73% |
| **DFTB** | **GFN1-xTB** | 5.18% | 11.88% | 11.24% | 26.51% | 18.08% | 18.12% | 17.60% |
| | **SCC-DFTB** | 41.05% | 37.01% | 31.37% | 13.11% | 29.94% | 8.19% | 31.63% |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 16.40% | 9.92% | 20.95% | 32.53% | 20.18% | 20.39% | 12.89% |
| | **PM7/B3LYP$_g$** | 14.68% | 20.02% | 25.03% | 32.25% | 24.83% | 22.37% | 14.71% |
| | **AM1/B3LYP$_s$** | 14.27% | 1.74% | 15.53% | 17.37% | 17.36% | 14.20% | 20.50% |
| | **PM7/B3LYP$_s$** | 18.71% | 10.03% | 20.75% | 17.62% | 19.23% | 15.03% | 18.37% |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 15.69% | 18.94% | 24.41% | 28.70% | 19.32% | 20.31% | 18.50% |
| | **SCC-DFTB/B3LYP$_g$** | 13.36% | 7.35% | 26.52% | 29.01% | 19.21% | 20.61% | 22.95% |
| | **GFN1-xTB/B3LYP$_s$** | 20.01% | 13.42% | 23.32% | 13.46% | 17.97% | 15.42% | 22.16% |
| | **SCC-DFTB/B3LYP$_s$** | 9.58% | 6.72% | 25.69% | 13.05% | 17.78% | 17.26% | 25.26% |

**Table S5.** $R^2$ of $\Delta E_r$ as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.99 | 0.36 | 0.98 | 0.83 | 0.56 | 0.69 | 0.72 |
| | **B3LYP$_g$** | 0.99 | 0.53 | 0.95 | 0.68 | 0.54 | 0.74 | 0.73 |
| | **PBE-D3$_g$** | 0.99 | 0.40 | 0.98 | 0.75 | 0.56 | 0.75 | 0.73 |
| | **B3LYP-D3$_g$** | 0.99 | 0.54 | 0.95 | 0.68 | 0.53 | 0.73 | 0.73 |
| | **PBE$_s$** | 0.94 | 0.99 | 0.98 | 0.89 | 0.64 | 0.87 | 0.70 |
| | **B3LYP$_s$** | 0.93 | 0.98 | 0.99 | 0.86 | 0.73 | 0.84 | 0.72 |
| | **PBE-D3$_s$** | 0.96 | 0.99 | 0.99 | 0.89 | 0.74 | 0.87 | 0.70 |
| | **B3LYP-D3$_s$** | 0.96 | 0.99 | 0.99 | 0.83 | 0.77 | 0.88 | 0.73 |
| **SEQM** | **AM1** | 0.70 | 0.72 | 0.89 | 0.78 | 0.53 | 0.86 | 0.71 |
| | **MNDO** | 0.62 | 0.00013 | 0.015 | 0.92 | 0.52 | 0.26 | 0.041 |
| | **MNDOD** | 0.94 | 0.00041 | 0.076 | 0.92 | 0.61 | 0.61 | 0.12 |
| | **PM3** | 0.32 | 0.18 | 0.34 | 0.94 | 0.22 | 0.75 | 0.59 |
| | **PM6** | 0.96 | 0.27 | 0.82 | 0.87 | 0.55 | 0.82 | 0.78 |
| | **PM6-D3H4X** | 0.96 | 0.27 | 0.82 | 0.87 | 0.55 | 0.82 | 0.78 |
| | **PM7** | 0.97 | 0.35 | 0.78 | 0.83 | 0.61 | 0.79 | 0.80 |
| **DFTB** | **GFN1-xTB** | 0.996 | 0.21 | 0.81 | 0.89 | 0.70 | 0.93 | 0.54 |
| | **SCC-DFTB** | 0.72 | 0.13 | 0.51 | 0.93 | 0.062 | 0.36 | 0.38 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.97 | 0.37 | 0.96 | 0.84 | 0.60 | 0.82 | 0.69 |
| | **PM7/B3LYP$_g$** | 0.89 | 0.38 | 0.95 | 0.87 | 0.37 | 0.73 | 0.74 |
| | **AM1/B3LYP$_s$** | 0.99 | 0.81 | 0.97 | 0.91 | 0.73 | 0.94 | 0.67 |
| | **PM7/B3LYP$_s$** | 0.16 | 0.72 | 0.96 | 0.89 | 0.42 | 0.88 | 0.77 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.92 | 0.45 | 0.96 | 0.88 | 0.53 | 0.79 | 0.75 |
| | **SCC-DFTB/B3LYP$_g$** | 0.96 | 0.63 | 0.96 | 0.89 | 0.59 | 0.83 | 0.67 |
| | **GFN1-xTB/B3LYP$_s$** | 0.046 | 0.41 | 0.93 | 0.92 | 0.61 | 0.93 | 0.74 |
| | **SCC-DFTB/B3LYP$_s$** | 0.36 | 0.45 | 0.74 | 0.91 | 0.81 | 0.89 | 0.41 |

**Table S6.** $R^2$ of LUMO energy of the reactant molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.98 | 0.93 | 0.90 | 0.78 | 0.67 | 0.86 | 0.64 |
| | **B3LYP$_g$** | 0.98 | 0.97 | 0.92 | 0.77 | 0.67 | 0.88 | 0.63 |
| | **PBE-D3$_g$** | 0.97 | 0.93 | 0.90 | 0.78 | 0.66 | 0.86 | 0.64 |
| | **B3LYP-D3$_g$** | 0.98 | 0.97 | 0.92 | 0.77 | 0.67 | 0.88 | 0.63 |
| | **PBE$_s$** | 0.995 | 0.99 | 0.91 | 0.80 | 0.67 | 0.91 | 0.55 |
| | **B3LYP$_s$** | 0.99 | 0.98 | 0.96 | 0.82 | 0.70 | 0.93 | 0.59 |
| | **PBE-D3$_s$** | 0.99 | 0.98 | 0.91 | 0.80 | 0.67 | 0.91 | 0.55 |
| | **B3LYP-D3$_s$** | 0.98 | 0.98 | 0.96 | 0.83 | 0.69 | 0.94 | 0.59 |
| **SEQM** | **AM1** | 0.98 | 0.97 | 0.75 | 0.50 | 0.71 | 0.69 | 0.53 |
| | **MNDO** | 0.96 | 0.62 | 0.76 | 0.36 | 0.62 | 0.73 | 0.44 |
| | **MNDOD** | 0.98 | 0.89 | 0.80 | 0.36 | 0.68 | 0.73 | 0.43 |
| | **PM3** | 0.996 | 0.57 | 0.66 | 0.53 | 0.81 | 0.67 | 0.51 |
| | **PM6** | 0.96 | 0.93 | 0.74 | 0.50 | 0.74 | 0.64 | 0.48 |
| | **PM6-D3H4X** | 0.98 | 0.93 | 0.74 | 0.50 | 0.70 | 0.64 | 0.48 |
| | **PM7** | 0.98 | 0.90 | 0.74 | 0.52 | 0.72 | 0.64 | 0.50 |
| **DFTB** | **GFN1-xTB** | 0.91 | 0.97 | 0.75 | 0.82 | 0.65 | 0.73 | 0.53 |
| | **SCC-DFTB** | 0.99 | 0.15 | 0.79 | 0.86 | 0.47 | 0.64 | 0.42 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.90 | 0.94 | 0.90 | 0.77 | 0.67 | 0.91 | 0.66 |
| | **PM7/B3LYP$_g$** | 0.97 | 0.999 | 0.94 | 0.75 | 0.69 | 0.92 | 0.67 |
| | **AM1/B3LYP$_s$** | 0.97 | 0.90 | 0.93 | 0.82 | 0.69 | 0.95 | 0.60 |
| | **PM7/B3LYP$_s$** | 0.94 | 0.97 | 0.94 | 0.79 | 0.72 | 0.94 | 0.65 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.98 | 0.95 | 0.93 | 0.76 | 0.64 | 0.89 | 0.65 |
| | **SCC-DFTB/B3LYP$_g$** | 0.97 | 0.99 | 0.95 | 0.86 | 0.69 | 0.91 | 0.60 |
| | **GFN1-xTB/B3LYP$_s$** | 0.85 | 0.97 | 0.96 | 0.81 | 0.67 | 0.94 | 0.63 |
| | **SCC-DFTB/B3LYP$_s$** | 0.94 | 0.93 | 0.98 | 0.91 | 0.70 | 0.95 | 0.55 |
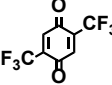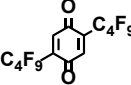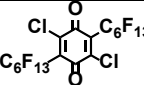
**Table S7.** $R^2$ of HOMO energy of the product molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.
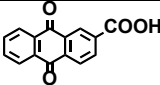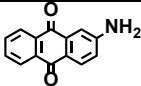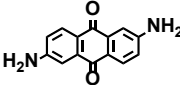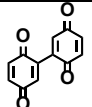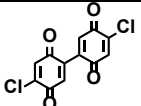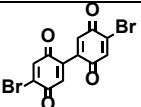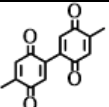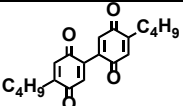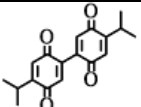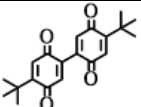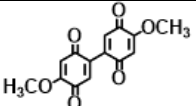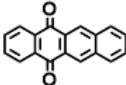
| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.58 | 0.46 | 0.38 | 0.51 | 0.16 | 0.65 | 0.80 |
| | **B3LYP$_g$** | 0.54 | 0.84 | 0.39 | 0.030 | 0.61 | 0.63 | 0.69 |
| | **PBE-D3$_g$** | 0.58 | 0.45 | 0.38 | 0.42 | 0.15 | 0.75 | 0.80 |
| | **B3LYP-D3$_g$** | 0.54 | 0.88 | 0.40 | 0.030 | 0.61 | 0.82 | 0.69 |
| | **PBE$_s$** | 0.85 | 0.44 | 0.72 | 0.77 | 0.43 | 0.73 | 0.72 |
| | **B3LYP$_s$** | 0.87 | 0.87 | 0.50 | 0.61 | 0.77 | 0.88 | 0.61 |
| | **PBE-D3$_s$** | 0.86 | 0.40 | 0.72 | 0.73 | 0.41 | 0.82 | 0.73 |
| | **B3LYP-D3$_s$** | 0.88 | 0.89 | 0.50 | 0.61 | 0.78 | 0.88 | 0.59 |
| **SEQM** | **AM1** | 0.78 | 0.93 | 0.22 | 0.67 | 0.68 | 0.75 | 0.71 |
| | **MNDO** | 0.90 | 0.059 | 0.17 | 0.83 | 0.45 | 0.75 | 0.50 |
| | **MNDOD** | 0.48 | 0.024 | 0.069 | 0.80 | 0.57 | 0.62 | 0.11 |
| | **PM3** | 0.90 | 0.80 | 0.11 | 0.83 | 0.76 | 0.81 | 0.42 |
| | **PM6** | 0.95 | 0.75 | 0.083 | 0.59 | 0.50 | 0.85 | 0.58 |
| | **PM6-D3H4X** | 0.94 | 0.74 | 0.081 | 0.59 | 0.47 | 0.85 | 0.58 |
| | **PM7** | 0.90 | 0.91 | 0.14 | 0.37 | 0.38 | 0.74 | 0.68 |
| **DFTB** | **GFN1-xTB** | 0.98 | 0.90 | 0.88 | 0.36 | 0.69 | 0.73 | 0.75 |
| | **SCC-DFTB** | 0.019 | 0.0037 | 0.081 | 0.84 | 0.14 | 0.95 | 0.18 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.84 | 0.93 | 0.59 | 0.039 | 0.61 | 0.66 | 0.86 |
| | **PM7/B3LYP$_g$** | 0.87 | 0.71 | 0.42 | 0.055 | 0.41 | 0.59 | 0.82 |
| | **AM1/B3LYP$_s$** | 0.88 | 0.998 | 0.77 | 0.73 | 0.71 | 0.83 | 0.65 |
| | **PM7/B3LYP$_s$** | 0.80 | 0.93 | 0.60 | 0.72 | 0.64 | 0.81 | 0.72 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.86 | 0.74 | 0.44 | 0.25 | 0.64 | 0.66 | 0.72 |
| | **SCC-DFTB/B3LYP$_g$** | 0.90 | 0.96 | 0.34 | 0.24 | 0.65 | 0.65 | 0.57 |
| | **GFN1-xTB/B3LYP$_s$** | 0.77 | 0.87 | 0.49 | 0.84 | 0.69 | 0.80 | 0.60 |
| | **SCC-DFTB/B3LYP$_s$** | 0.95 | 0.97 | 0.38 | 0.85 | 0.70 | 0.76 | 0.48 |

**Table S8.** Average NRMSE of the seven datasets for LUMO descriptor as calculated with B3LYP, SEQM, DFTB, and the hybrid schemes of SEQM (or DFTB) and B3LYP.

| Methods | Average NRMSE | Methods | Average NRMSE |
|---|---|---|---|
| $B3LYP_g$ | 12.65% | $AM1/B3LYP_g$ | 13.83% |
| AM1 | 16.10% | $PM7/B3LYP_g$ | 11.46% |
| MNDO | 19.61% | $AM1/B3LYP_s$ | 12.76% |
| MNDOD | 17.50% | $PM7/B3LYP_s$ | 12.25% |
| PM3 | 18.17% | $GFN1\text{-}xTB/B3LYP_g$ | 12.99% |
| PM6 | 17.27% | $SCC\text{-}DFTB/B3LYP_g$ | 11.60% |
| PM6-D3H4X | 17.16% | $GFN1\text{-}xTB/B3LYP_s$ | 13.11% |
| PM7 | 17.13% | $SCC\text{-}DFTB/B3LYP_s$ | 12.01% |
| GFN1-xTB | 15.85% | | |
| SCC-DFTB | 19.60% | | |

**Table S9.** Summary of operation voltage and charge/discharge windows for the seven datasets.

| Dataset | Molecular structure | Operation voltage (vs. Li/Li$^+$) | Charge/discharge window (vs. Li/Li$^+$) |
|---|---|---|---|
| 1 | | 2.5 | 1.2 – 4.3 |
| | | 3.0 | |
| | | 3.1 | |
| | | 3.1 | |
| 2 | | 2.61 | Not given |
| | | 2.52 | |
| | | 2.71 | |
| | | 2.27 | |
| 3 | | 2.61 | |
| | | 2.68 | |
| | | 2.74 | |
| | | 2.93 | |
| | | 2.27 | |
| 4 | | 3.28 | 1.5 – 4.0 |
| | | 2.35 | |
| | | 2.01 | |

| Group | Structure | Value | Range |
|---|---|---|---|
| | (anthraquinone-2-carboxylic acid) | 2.04 | |
| | (2-aminoanthraquinone) | 1.86 | |
| | (2,6-diaminoanthraquinone) | 1.73 | |
| **5** | (bi(benzoquinone)) | 2.8 | 1.5 – 4.2 |
| | (dichloro bi(benzoquinone)) | 2.8 | |
| | (dibromo bi(benzoquinone)) | 2.9 | |
| | (dimethyl bi(benzoquinone)) | 2.6 | |
| | (di-$C_4H_9$ bi(benzoquinone)) | 2.7 | |
| | (diisopropyl bi(benzoquinone)) | 2.7 | |
| | (di-tert-butyl bi(benzoquinone)) | 2.6 | |
| | (dimethoxy bi(benzoquinone)) | 2.7 | |
| **6** | (1,4-naphthoquinone) | 2.4 | 1.5 – 3.5 |
| | (anthraquinone) | 2.1 | |
| | (naphthacenequinone) | 1.7 | |
| | (phenanthrenequinone) | 2.5 | |

| | | | |
|---|---|---|---|
| | (structure) | 2.7 | |
| **7** | (structure) | 1.92 | |
| | (structure) | 1.79 | |
| | (structure) | 2.53 | |
| | (structure) | 2.11 | |
| | (structure) | 2.32 | |
| | (structure) | 2.39 | |
| | (structure) | 2.61 | |

**Table S10.** The 2D structures of seven quinones, and zero-point energy ($ZPE$) of quinone reactants and lithiated products and their difference on $ZPE$, as calculated with the functionals of PBE and B3LYP in gas phase.
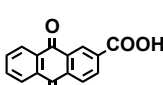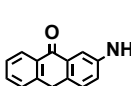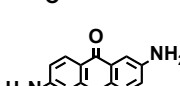
| 2D molecules | PBE | | | B3LYP | | |
|---|---|---|---|---|---|---|
| | *ZPE* for reactant (eV) | *ZPE* for product (eV) | *Δ(ZPE)* (eV) | *ZPE* for reactant (eV) | *ZPE* for product (eV) | *Δ(ZPE)* (eV) |
|  | 2.25 | 2.32 | 0.07 | 2.32 | 2.36 | 0.04 |
|  | 3.51 | 3.56 | 0.05 | 3.61 | 3.69 | 0.08 |
|  | 4.77 | 4.81 | 0.04 | 4.91 | 4.92 | 0.01 |
|  | 5.16 | 5.20 | 0.04 | 5.31 | 5.36 | 0.05 |
|  | 5.19 | 5.27 | 0.08 | 5.33 | 5.41 | 0.08 |
|  | 5.61 | 5.71 | 0.10 | 5.76 | 5.86 | 0.10 |
|  | 4.70 | 4.83 | 0.13 | 4.84 | 4.96 | 0.12 |

**Table S11.** RMSE of $\Delta E_r$ as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.019 | 0.13 | 0.034 | 0.21 | 0.064 | 0.19 | 0.15 |
| | **B3LYP$_g$** | 0.019 | 0.11 | 0.047 | 0.29 | 0.066 | 0.18 | 0.15 |
| | **PBE-D3$_g$** | 0.019 | 0.13 | 0.034 | 0.26 | 0.064 | 0.17 | 0.15 |
| | **B3LYP-D3$_g$** | 0.023 | 0.11 | 0.049 | 0.29 | 0.066 | 0.18 | 0.15 |
| | **PBE$_s$** | 0.059 | 0.017 | 0.027 | 0.17 | 0.058 | 0.123 | 0.16 |
| | **B3LYP$_s$** | 0.064 | 0.021 | 0.019 | 0.19 | 0.050 | 0.14 | 0.15 |
| | **PBE-D3$_s$** | 0.050 | 0.018 | 0.026 | 0.17 | 0.049 | 0.12 | 0.16 |
| | **B3LYP-D3$_s$** | 0.052 | 0.020 | 0.018 | 0.21 | 0.047 | 0.12 | 0.15 |
| **SEQM** | **AM1** | 0.14 | 0.086 | 0.073 | 0.24 | 0.066 | 0.13 | 0.15 |
| | **MNDO** | 0.15 | 0.16 | 0.21 | 0.15 | 0.067 | 0.30 | 0.28 |
| | **MNDOD** | 0.060 | 0.16 | 0.21 | 0.15 | 0.060 | 0.22 | 0.27 |
| | **PM3** | 0.21 | 0.15 | 0.18 | 0.13 | 0.086 | 0.18 | 0.18 |
| | **PM6** | 0.051 | 0.14 | 0.092 | 0.19 | 0.065 | 0.15 | 0.14 |
| | **PM6-D3H4X** | 0.052 | 0.14 | 0.092 | 0.19 | 0.065 | 0.15 | 0.14 |
| | **PM7** | 0.040 | 0.13 | 0.11 | 0.21 | 0.060 | 0.16 | 0.13 |
| **DFTB** | **GFN1–xTB** | 0.016 | 0.15 | 0.095 | 0.17 | 0.053 | 0.093 | 0.19 |
| | **SCC-DFTB** | 0.13 | 0.15 | 0.15 | 0.13 | 0.094 | 0.28 | 0.23 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.045 | 0.13 | 0.046 | 0.21 | 0.061 | 0.15 | 0.16 |
| | **PM7/B3LYP$_g$** | 0.084 | 0.13 | 0.047 | 0.19 | 0.077 | 0.18 | 0.15 |
| | **AM1/B3LYP$_s$** | 0.020 | 0.072 | 0.038 | 0.16 | 0.050 | 0.085 | 0.16 |
| | **PM7/B3LYP$_s$** | 0.23 | 0.086 | 0.045 | 0.17 | 0.074 | 0.12 | 0.14 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.069 | 0.12 | 0.042 | 0.18 | 0.066 | 0.16 | 0.14 |
| | **SCC-DFTB/B3LYP$_g$** | 0.046 | 0.10 | 0.045 | 0.17 | 0.062 | 0.14 | 0.17 |
| | **GFN1-xTB/B3LYP$_s$** | 0.24 | 0.13 | 0.056 | 0.14 | 0.061 | 0.091 | 0.14 |
| | **SCC-DFTB/B3LYP$_s$** | 0.20 | 0.12 | 0.11 | 0.16 | 0.042 | 0.12 | 0.22 |

**Table S12.** RMSE of LUMO energy of the reactant molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.038 | 0.043 | 0.070 | 0.24 | 0.056 | 0.13 | 0.17 |
| | **B3LYP$_g$** | 0.031 | 0.029 | 0.059 | 0.25 | 0.056 | 0.12 | 0.17 |
| | **PBE-D3$_g$** | 0.040 | 0.043 | 0.070 | 0.24 | 0.057 | 0.13 | 0.17 |
| | **B3LYP-D3$_g$** | 0.033 | 0.029 | 0.060 | 0.25 | 0.056 | 0.12 | 0.17 |
| | **PBE$_s$** | 0.017 | 0.016 | 0.065 | 0.23 | 0.056 | 0.11 | 0.19 |
| | **B3LYP$_s$** | 0.030 | 0.023 | 0.044 | 0.22 | 0.053 | 0.093 | 0.18 |
| | **PBE-D3$_s$** | 0.025 | 0.022 | 0.063 | 0.23 | 0.056 | 0.11 | 0.19 |
| | **B3LYP-D3$_s$** | 0.033 | 0.024 | 0.044 | 0.21 | 0.054 | 0.087 | 0.18 |
| **SEQM** | **AM1** | 0.037 | 0.027 | 0.11 | 0.36 | 0.052 | 0.19 | 0.20 |
| | **MNDO** | 0.047 | 0.10 | 0.11 | 0.41 | 0.060 | 0.18 | 0.21 |
| | **MNDOD** | 0.036 | 0.055 | 0.097 | 0.41 | 0.055 | 0.18 | 0.22 |
| | **PM3** | 0.015 | 0.11 | 0.13 | 0.35 | 0.043 | 0.20 | 0.20 |
| | **PM6** | 0.048 | 0.043 | 0.11 | 0.37 | 0.050 | 0.21 | 0.21 |
| | **PM6-D3H4X** | 0.039 | 0.043 | 0.11 | 0.37 | 0.053 | 0.21 | 0.21 |
| | **PM7** | 0.038 | 0.051 | 0.11 | 0.36 | 0.051 | 0.21 | 0.20 |
| **DFTB** | **GFN1-xTB** | 0.075 | 0.030 | 0.11 | 0.22 | 0.057 | 0.18 | 0.20 |
| | **SCC-DFTB** | 0.026 | 0.15 | 0.099 | 0.19 | 0.070 | 0.21 | 0.22 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.077 | 0.039 | 0.067 | 0.25 | 0.055 | 0.10 | 0.17 |
| | **PM7/B3LYP$_g$** | 0.041 | 0.0060 | 0.051 | 0.26 | 0.054 | 0.098 | 0.16 |
| | **AM1/B3LYP$_s$** | 0.043 | 0.051 | 0.057 | 0.22 | 0.054 | 0.081 | 0.18 |
| | **PM7/B3LYP$_s$** | 0.059 | 0.030 | 0.052 | 0.23 | 0.051 | 0.084 | 0.17 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.039 | 0.036 | 0.057 | 0.25 | 0.058 | 0.11 | 0.17 |
| | **SCC-DFTB/B3LYP$_g$** | 0.045 | 0.016 | 0.048 | 0.19 | 0.054 | 0.10 | 0.18 |
| | **GFN1-xTB/B3LYP$_s$** | 0.096 | 0.030 | 0.043 | 0.22 | 0.055 | 0.084 | 0.17 |
| | **SCC-DFTB/B3LYP$_s$** | 0.061 | 0.043 | 0.034 | 0.15 | 0.053 | 0.079 | 0.19 |

**Table S13.** RMSE of HOMO energy of the product molecules as computed with DFT, SEQM, DFTB and hybrid schemes of SEQM (or DFTB) and DFT methods for the seven datasets.

| | | No.1 | No.2 | No.3 | No.4 | No.5 | No.6 | No.7 |
|---|---|---|---|---|---|---|---|---|
| **DFT** | **PBE$_g$** | 0.16 | 0.12 | 0.17 | 0.36 | 0.089 | 0.21 | 0.13 |
| | **B3LYP$_g$** | 0.17 | 0.065 | 0.17 | 0.51 | 0.061 | 0.21 | 0.16 |
| | **PBE-D3$_g$** | 0.16 | 0.12 | 0.17 | 0.39 | 0.089 | 0.18 | 0.13 |
| | **B3LYP-D3$_g$** | 0.17 | 0.058 | 0.17 | 0.51 | 0.060 | 0.15 | 0.16 |
| | **PBE$_s$** | 0.095 | 0.12 | 0.11 | 0.25 | 0.073 | 0.18 | 0.15 |
| | **B3LYP$_s$** | 0.089 | 0.059 | 0.15 | 0.32 | 0.047 | 0.12 | 0.18 |
| | **PBE-D3$_s$** | 0.092 | 0.13 | 0.11 | 0.27 | 0.074 | 0.15 | 0.15 |
| | **B3LYP-D3$_s$** | 0.086 | 0.055 | 0.15 | 0.32 | 0.045 | 0.12 | 0.18 |
| **SEQM** | **AM1** | 0.12 | 0.042 | 0.19 | 0.30 | 0.055 | 0.17 | 0.15 |
| | **MNDO** | 0.080 | 0.16 | 0.20 | 0.21 | 0.072 | 0.18 | 0.20 |
| | **MNDOD** | 0.18 | 0.16 | 0.21 | 0.23 | 0.063 | 0.22 | 0.27 |
| | **PM3** | 0.077 | 0.074 | 0.20 | 0.21 | 0.047 | 0.15 | 0.22 |
| | **PM6** | 0.054 | 0.082 | 0.21 | 0.33 | 0.068 | 0.14 | 0.19 |
| | **PM6-D3H4X** | 0.059 | 0.083 | 0.21 | 0.33 | 0.071 | 0.14 | 0.19 |
| | **PM7** | 0.077 | 0.048 | 0.20 | 0.41 | 0.076 | 0.18 | 0.16 |
| **DFTB** | **GFN1-xTB** | 0.031 | 0.052 | 0.074 | 0.41 | 0.054 | 0.18 | 0.14 |
| | **SCC-DFTB** | 0.25 | 0.16 | 0.21 | 0.20 | 0.090 | 0.082 | 0.26 |
| **SEQM/ DFT hybrid** | **AM1/B3LYP$_g$** | 0.098 | 0.044 | 0.14 | 0.50 | 0.061 | 0.20 | 0.11 |
| | **PM7/B3LYP$_g$** | 0.088 | 0.088 | 0.17 | 0.50 | 0.074 | 0.22 | 0.12 |
| | **AM1/B3LYP$_s$** | 0.086 | 0.0080 | 0.10 | 0.27 | 0.052 | 0.14 | 0.17 |
| | **PM7/B3LYP$_s$** | 0.11 | 0.044 | 0.14 | 0.27 | 0.058 | 0.15 | 0.15 |
| **DFTB/ DFT hybrid** | **GFN1-xTB/B3LYP$_g$** | 0.094 | 0.083 | 0.16 | 0.45 | 0.058 | 0.20 | 0.15 |
| | **SCC-DFTB/B3LYP$_g$** | 0.080 | 0.032 | 0.18 | 0.45 | 0.058 | 0.21 | 0.19 |
| | **GFN1-xTB/B3LYP$_s$** | 0.12 | 0.059 | 0.15 | 0.21 | 0.054 | 0.15 | 0.18 |
| | **SCC-DFTB/B3LYP$_s$** | 0.057 | 0.030 | 0.17 | 0.20 | 0.053 | 0.17 | 0.21 |

**References:**

S1.   Xu, K. Nonaqueous liquid electrolytes for lithium-based rechargeable batteries. *Chem. Rev.* **2004**, *104*, 4303–4418.

S2.   Prakongpan, S.; Nagai, T. Solubility of acetaminophen in cosolvents. *Chem. Pharm. Bull.* **1984**, *32*, 340–343.

S3.   Jouyban, A.; Soltanpour, S. Prediction of dielectric constants of binary solvents at various temperatures. *J. Chem. Eng. Data* **2010**, *55*, 2951–2963.