

Information for the Dataset Provided

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Introduction

This document describes the dataset provided. The dataset is for the publication: “Development of a multi-step screening procedure for redox active molecules in organic radical polymer anodes and as redox flow anolytes” by Andreas J. Achazi, Xhesilda Fataj, Philip Rohland, Martin D. Hager, Ulrich S. Schubert, Doreen Mollenhauer (<https://doi.org/10.1002/jcc.27299>). The folders usually contain subfolders. The folder structure indicates the heteroatom, the linker R, the level of theory and the charge of the compound. GB3LYP refers to B3LYP where the VWN3 density functional is used instead of the VWN5 density functional.

Furthermore, n, p and m suffixes are also used for the folder names. “n” stands for neutral compound, “p” stands for plus, which means the compound has a charge of +1, and “m” stands for minus, meaning the compound has a charge of -1.

Computational details

Semi-empirical Methods

The calculations at the xTB level of theory were performed using the CREST program package.¹⁻³ The extended tight-binding model GFN2-xTB⁴⁻⁶ was applied. The solvent acetonitrile was simulated in these calculations using the implicit analytical linearised Poisson-Boltzmann (ALPB) and generalised Born with surface area (GBSA) solvent models.⁷

Density Functional Theory (DFT)

DFT calculations were performed using the Turbomole 7.5.1 software package.⁸⁻¹⁰ In some cases the Gaussian 16 C.01 program package was used¹¹ when the density functionals (DFs) or solvent model

were not available in Turbomole 7.5.1. When Turbomole 7.5.1 was used, the calculations were speeded up by the Resolution-of-Identity (RI) approximation.¹²⁻¹⁴ Furthermore, the “multiple grid” m4 was utilised.¹⁵ When B3LYP-D3(BJ)¹⁶⁻²³ (COSMO)^{24,25} and BP86-D3(BJ)²⁶ (COSMO) were used, the convergence criteria were set to $10^{-8} E_h$ and $10^{-4} E_h a_0^{-1}$ for the energy and gradient. For all other DFs used in Turbomole 7.5.1 the convergence was set to $10^{-6} E_h$ and $10^{-3} E_h a_0^{-1}$, respectively. The solvent acetonitrile (relative permittivity $\epsilon_r = 37.5$,²⁷ refractive index $n_D = 1.3442$ ²⁸), was simulated with different implicit solvent models: COSMO, COSMO-out,²⁹ COSMO-ion,³⁰ COSMO-ion-out, DCOSMO-RS,³¹ DCOSMO-RS-out and DCOSMO-RS-out-c. In addition to D3(BJ), the dispersion corrections D4^{32,33} and VV10^{34,35} were applied.

The DFs: LDA [=SVWN(V)],¹⁶⁻¹⁸ B3LYP-D3(BJ),¹⁶⁻²³ BHLYP-D3(BJ),³⁶ CAM-B3LYP-D3(BJ),³⁷ BP86-D3(BJ),²⁶ M06-L,³⁸ M06,³⁸ M06-2X,³⁸ M11-L,³⁹ M11,⁴⁰ revM11,⁴¹ MN12-L,⁴² MN12-SX,⁴³ MN15-L,⁴⁴ MN15,⁴⁵ PBE-D3(BJ),^{46,47} PBE0-D3(BJ),⁴⁸ TPSS-D3(BJ),⁴⁹ TPSSH-D3(BJ),⁵⁰ PW6B95-D3(BJ),⁵¹ r2SCAN-D4,⁵² SCAN-D3(BJ),⁵³ and SCAN0⁵⁴ were used. Calculations were performed using the def2-XVP (X = S, QZ),^{14,15,55} def2-SVPD^{14,56} and def2-XVPPD (X = TZ, QZ)^{14,56} basis sets. In addition, PBEh-3c/def2-mSVP,⁵⁷ r2SCAN-3c/mTZVPP,⁵⁸ and HF-3c/minix⁵⁹ calculations were performed. HF-3c is wave function based and not DFT based.

M08-HX,⁶⁰ wB97X⁶¹ and wB97XD⁶² were performed with the IEFPCM^{63,64} solvent model and without RI approximation using the Gaussian 16 C.01 program package. Furthermore, B3LYP-D3(BJ) calculations were also performed with the IEFPCM and the Solvation Model based on Density (SMD)⁶⁵ using Gaussian 16 C.01. The B3LYP DF in Gaussian 16 C.01 utilizes the VWN3 functional, while Turbomole 7.5.1 utilizes the VWN5 functional. Furthermore, for solvent models in Gaussian 16 C.01 the relative permittivity $\epsilon_r = 35.688$ for acetonitrile is used. For comparison, calculations in Turbomole 7.5.1 were performed using VWN3 for B3LYP and $\epsilon_r = 35.688$.

Radial Grid Sizes for SCAN DF

The radial grid size (radsize) was increased to 8 (in TURBOMOLE notation) for r2SCAN-D4 and r2SCAN-3c, and to 14 for SCAN-D3(BJ) and SCAN0. A radial grid size of 20 was also applied. For all other DFs in Turbomole 7.5.1 a radial grid size of 6 was used.

Reparametrizing DFs

The Hartree-Fock exchange of B3LYP-D3(BJ)⁺, B3LYP-D3(BJ)⁻, PBE0-D3(BJ)⁺ and PBE0-D3(BJ)⁻ was set to 20.49%, 28.04%, 16.05%, and 24.83%, respectively. Otherwise, these density functionals are identical to the B3LYP-D3(BJ) and PBE0-D3(BJ) DFs. Furthermore, B3LYP-D3(BJ)/def2-SVP and PBE0-D3(BJ)/def2-SVP calculations were performed with Hartree-Fock exchanges from 5% to 50% ($c = 0.5$) in 1% steps.

Other Parameters

For the other parameters, the values predefined in Turbomole 7.5.1 or Gaussian 16 C.01 were selected.

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