EDLC materials design using Ab-initio Molecular Orbital Calculations

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Organic electrolytes have been widely studied as basic materials for electrochemical devices such as a rechargeable battery and an electric double layer capacitor (EDLC)[1]. For an EDLC, the operating voltage determine their device size, energy density and long term durability. Computational chemistry has been thought as a potential EDLC materials design tool. However, precise estimation of redox potentials of electrolytes has not been established.

In this study, EDLC materials design using ab-initio molecular orbital calculations under Hartree-Fock level of theory and 3-21+G basis set has been discussed. The cation-anion and solute-solvent interactions by embedding an cation-anion cluster in cavity of dielectric continuum using isodensity polarizable continuum model (IPCM)[2] have been taken into account. Figure 1 shows comparison between theoretical and experimental value of redox potentials of several types of electrolytes consist from six cation listed in Fig. 2 and tetrafuloroborate (BF_4^-). Propylene carbonate (PC) was used as a solvent for all cases. The theoretical value well reproduced their experimental values. The intermolecular interactions between cation and anion can play an important role for the redox potential windows of the EDLC electrolytes.

Ab-initio molecular orbital calculations can be a powerful tool for EDLC material design if precise inter molecular interactions were taken into account.

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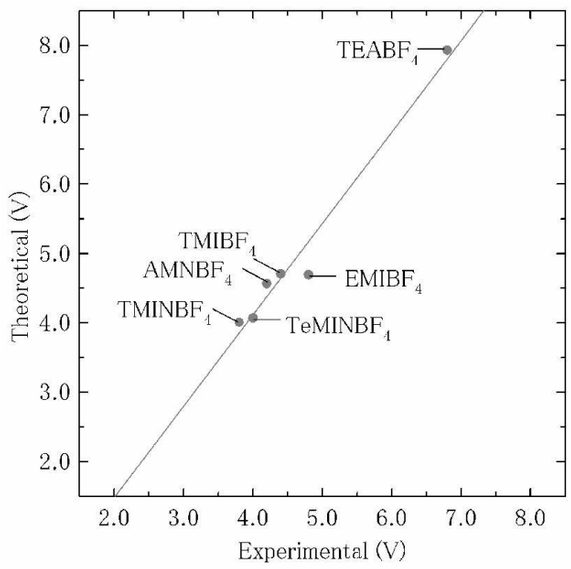

Fig. 1. Comparison between theoretical and experimental values of redox potential windows of electrolytes. Theoretical calculations with precise inter molecular interactions well reproduced their experimental values.

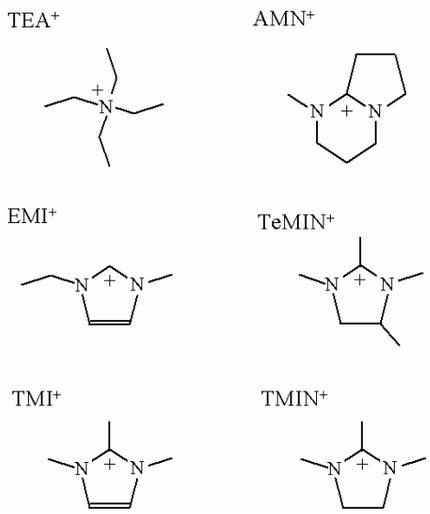

Fig.2. Chemical formulas of cations used in this study.