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Materials for Energy by Computational Design: exploring organic electrodes toward next-generation batteries

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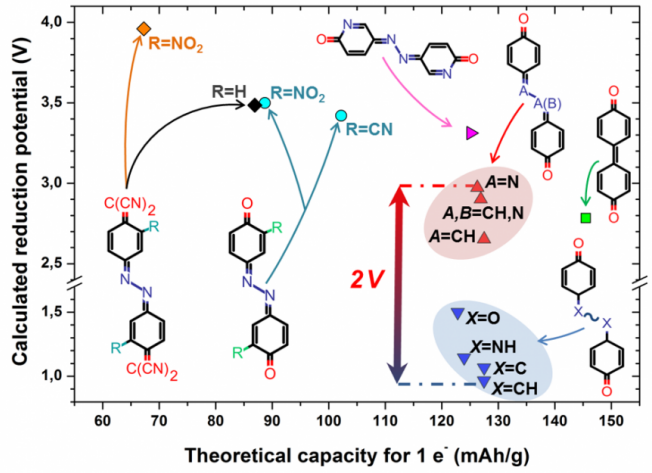
PURPOSE OF THE ABSTRACT

In recent years, the demand for sustainable energy sources has stimulated great interest in new technologies and materials. A multitude of research efforts have led to marked progress in this area, including in particular Lithium-ion batteries based on organic electrodes. A significant milestone in designing such materials would be achieved by reaching the complete comprehension of the rules governing the increase/decrease of the redox potential of molecular entities, especially if differentiation between various backbones/functional groups/redox centres or incidence of atom substitution within rings can be reached. Based on the combined use of molecular quantum simulation and accurate examination of the electronic structure properties, a rational design of organic electrode materials is reachable. In particular, we developed an approach based on the consideration of various indicators extracted from the geometry/electronic structure (HOMA (Harmonic Oscillator Model of Aromaticity), delocalization indices such as FLU parameter), which give the opportunity to appreciate electron delocalization on various rings/bond paths within a molecule in a quantitative way. In this context, we also evidenced that the partitioning of the global energy of the molecule constitutes another analysis instrument to shed light on the various pieces of molecule playing the main role in stabilization/destabilization upon reduction or how balance between some competitive effects does occur. We also focused on plots of electrostatic potential (ESP) on the molecular surface which give meaningful insight into charge distributions in molecules, these color-coded maps allowing in particular to visualize the preferred sites of Li+ positioning around the molecular ion [1]. Modelling of energy materials may advantageously complete the experimental studies not only through the prediction of already existing molecules and materials simply tuned through functionalization/isomerism and substitution but also thanks to the suggestion of new chemical engineering (new backbones) originating from computational design. Such global approach can be applied to systematic theoretical screening of existing compounds/hypothetical novel candidates, both being tuned through various effects. This should progressively complete the database and pave the way toward the discovery of new/optimized organic electrodes with improved features. Thanks to this methodology a work of prospection has been undertaken on various families, including derivatives of quinone, carboxylate, quinoneazine and pentalenedione [1-7]. By focusing on these various sets of compounds, we identified some property-based guidelines, which may serve not only for the ranking of the studied entities by themselves but also for the search of more advanced and innovative systems.

FIGURES



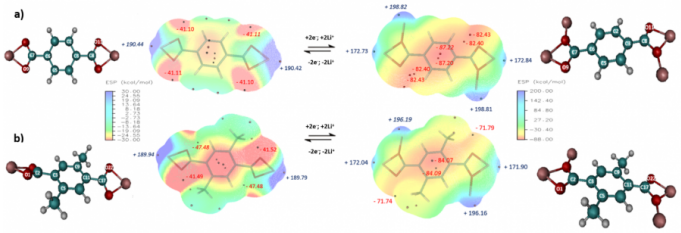


FIGURE 1

Figure 1

Calculated reduction potential (in V vs. Li+/Li) as a function of the capacity for a one-electron process in quinoneazine-derivatives.

FIGURE 2

Figure 2

DFT-optimized conformations (stick and balls representation) and electrostatic maps of compound (a) dilithium terephthalate, (b) dilithium dimethyl terephthalate before (on the left) and after double reduction (on the right).

KEYWORDS

organic electrode material | computation design and quantum chemical modelling | chemical engineering | electronic structure

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