Automated Generation and Theoretical Predictions for Potential Near-Infrared (NIR) Dye Sensitized Solar Cells: Generating Theoretical Dyes

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Abstract

Dye-sensitized solar cells (DSCs) use organic sensitizers to absorb light, making them a cheaper and environmentally less toxic option than current silicon based solar cells. The sensitizing dyes can be tuned to absorb the sunlight's photons, enabling dyes to be stacked within a DSC to improve the entire DSC's efficiency. With many limitations in costs to experimentally construct a database of these dyes for finding the best combinations of these dyes, the usage of time-dependent density functional theory (TD-DFT) can provide vertical transition energy predictions to proactively investigate dyes before investing in their synthesis. Additionally, through using a triple donor method that has an electron acceptor, backbone, and electron donor piece to generate new structures, theoretical work has generated a dye dataset of over 2000 structures. The structures are optimized with B3LYP/6-311G(d,p) before calculating electronic excited states with CAM-B3LYP/6-311G(d,p), BHandHLYP/6-311G(d,p) and PBE0/6-311G(d,p). The excitation energies map to the experimental λ_{max} values with CAM-B3LYP behaving as an upper bound for 86.1% of the 72 benchmark dyes and PBE0 as a minimum for 93.1% of dyes. Through combining the CAM-B3LYP/6-311G(d,p) and PBE0/6-311G(d,p) energies through using a least squares fitting (LSF) approach, a prediction for the λ_{max} has a mean absolute error of 0.13 eV. The computed LUMO energies are correlated to experimental LUMO values through the LSF approach to acquire an root mean square error of 0.11 eV enabling experimentalists to have more confidence in these theoretical dyes performing well in a DSC beyond just having the right absorption energy. The theoretical dataset consists of absorption energies ranging from 1.61 eV to 3.56 eV producing candidate DSC's with a maximum efficiency of up to 33%. Ultimately, this computational approach will expedite the search for the perfect

combination of dyes to beat silicon solar cells through both a better price point and efficiency.