Abstract

Over the last few years, organic solar cells (OSCs) have emerged as one of the most promising third-generation photovoltaic technologies due to unique advantages like low production costs, roll-to-roll solution processability, lightweight, and mechanical flexibility. Recent developments in novel conjugated polymer donor and non-fullerene acceptor (NFA) materials with promising properties have led to an unprecedented increase in power conversion efficiency (PCE) by more than 20%. Further improvement is still attainable with the optimal combinations of donors and acceptors that provide minimal voltage and current losses. However, in this era of artificial intelligence, identifying the highly potential combinations of such donor and acceptor materials using the current trial-and-error experimental approaches is certainly not feasible.

In this thesis, we explored the data-driven machine-learning (ML) framework for the analysis and prediction of photovoltaic parameters in non-fullerene organic solar cells and successively validated this predictivity by fabricating a set of highly efficient devices. Firstly, the data-enabled ML framework was employed to predict the energy losses using frontier molecular orbital (FMO) descriptors in the polymer:NFA-based devices. The Random Forest regression model showed the best performance in predicting the energy losses, with a correlation coefficient of 0.83 and a relative error in the range of 0–20%. We extended our work to predicting photovoltaic parameters (short circuit current density, open circuit voltage and PCE) using various combinations of FMO and RDKit descriptors and then validated the model by fabricating the devices. A dataset of 1242 experimentally verified donor:acceptor combinations was constructed, and the corresponding material descriptors were generated to train and test five different supervised ML models. Using a unique combination of both FMO and RDKit descriptors as input features, the random forest ML model performed best for predicting the PCE with a Person’s coefficient of 0.791 and a mean absolute percentage error

of 2.004. Furthermore, the importance of critical RDKit descriptors along with FMO descriptors in such performance predictions was realized by SHapley Additive exPlanations (SHAP) analyses. Therefore, these new descriptors will guide the proposed ML framework, which will be beneficial for designing new molecules, screening, and predicting suitable donor:acceptor combinations, thereby accelerating the development of highly efficient OSCs.

In addition to the donor:acceptor virtual screening and performance predication, OSCs still have a large open circuit voltage loss (ΔV_{OC} ~ 0.6 V) because of molecular disorder and high energetic offsets, as well as photocurrent losses because of the excitonic nature of photoactive layer. For this, we explored how exciton lifetime, energetic offsets, and disorder affect the voltage loss in bulk heterojunction organic solar cells. This has enabled us to identify the key features for minimizing the voltage loss, like (1) a low energy offset between the donor and acceptor molecular states is essential to attain a non-radiative voltage loss as low as ~200 meV and (2) Urbach energy, which is a measure of the materials' disorder and packing, should be low for the minimization of the radiative voltage loss.

On the other hand, the photocurrent losses were reduced by utilizing the synergistic plasmonic effects of multi-shaped Au nanostructures (diameter/edge length ~ 50 nm) hybridized with few-layer WS_2 nanosheets to improve the photocurrent of fullerene and non-fullerene-based OSCs. A PCE enhancement of more than 15% and an external quantum efficiency improvement in a broad wavelength range of 350-700 nm were demonstrated by incorporating these Au-WS_2 nanohybrids as an interlayer between hole-transport and photoactive layers. The effective plasmonic range over 350–700 nm and more than 50% forward light scattering indicated that these hybridized Au nanostructures are highly suitable candidates to couple the incident light into the active layer. Near field intensity enhancement and subsequent enhancement in absorption density confirmed the elevated local density of optical states in Au-WS_2 nanohybrid-based devices. Therefore, the present study emphasizes

the role of hybridized Au-WS_2 nanostructures in improving the light-harvesting capability of OSCs in a broad wavelength range, thereby attaining a significant improvement in device photocurrent. Overall, this thesis helps to screen the potential donor:acceptor combination and how to reduce photovoltage and photocurrent losses in OSCs for its successful commercialization.