Sheet-To-Sheet and Roll-To-Roll Processing of Large-Area Organic Photovoltaic Cells, Modules, and Tandem Devices

Abstract

The field of organic photovoltaics has existed for a few decades and is regarded as an environmentally friendly energy technology that could lower the carbon footprint. Recently, the development of new materials has led to power conversion efficiency (PCE) exceeding 20 %, revitalizing the field. However, almost all state-of-the-art devices reaching these performance values are fabricated via spin-coating techniques. However, for commercial success, an alternative industry-friendly Roll-to-Roll (R2R) compatible technique, such as slot-die coating, should be employed to scale up the devices, along with layers processed from greener solvents.

In this thesis, we focused on developing high-performing and stable organic photovoltaics using industry-compatible scale-up techniques such as Sheet-to-Sheet (S2S) slot-die and Roll-to-Roll sputtering. These devices were designed to serve various applications; prototypes were developed to demonstrate their potential. First, we demonstrated high-performance devices by formulating greener solvent inks, achieving high-quality layer formation, and establishing proper interconnections. Here, the slot-die technique was employed to process a few critical layers of the devices, namely, the electron transport layer, the hole transport layer, and the photoactive layer. For the photoactive layer, bulk heterojunction (BHJ) and Layer-by-Layer (LBL) film formation strategies were employed. For the best-performing devices, power conversion efficiencies of 15.24 % for BHJ and 14.10 % for LBL were achieved in small-cell devices, respectively. A detailed thermal degradation study (ISOS-D1) of devices with inverted and conventional configurations, employing photoactive layers via BHJ and LBL, revealed that inverted devices are more stable than conventional devices. The morphological study showed that the top electrode and the top transport layers are the main pathways of degradation. Additionally, a PCE of 13.02 % on mini-modules, measured under one sun, was achieved with the PM6 donor and Y7-12 acceptor. Furthermore, these devices maintained stable performance for more than 800 hours when continuously illuminated under one-sun light with a long-pass filter.

To further exploit the scalable technique, devices were developed for indoor applications where high-quality thin films are in demand. Here, devices were fabricated to achieve layers with fewer pinholes, thereby obtaining extremely low dark-leakage current and enabling operation under lower luminosities. The devices were fabricated using slot die coating iii

techniques, as stated earlier. In a mini-module, an efficiency of 12.62 % was achieved using the PM6 donor and the Y7-12 acceptor. When a wide-band-gap polymer donor, PTQ10, was blended with a medium band-gap acceptor, FCC-Cl, the efficiencies of small cells increased to a record high of 23.4 %, and for mini-modules to 19.65 %, measured at 1000 lux using an 2100 k LED lamp.

To further improve the scalability of the devices, we employed techniques that are fully compatible with R2R processing. Here, we combined R2R vacuum sputtering and S2S solution coating slot-die methods as a hybrid process to establish a complete industry-scale process. We developed a smooth silver bottom electrode using the R2R vacuum sputtering technique, which facilitates the formation of other layers on top. The fabricated mini-modules demonstrated a performance of 12.5 % PCE on a glass substrate. When the process was transferred to a PET substrate measuring 24 cm × 17 cm, the mini-modules exhibited an average PCE of 11.2 %, with the highest value reaching 11.5 %. A prototype mobile charger was developed to demonstrate the potential for scalability in practical applications. It achieved a record-high efficiency of 9.9 % on an active area >100 cm2, fabricated on a flexible PET substrate in an ITO-free configuration using fully R2R-compatible processing.

Finally, as part of the H2020 project CITYSOLAR, Tandem transparent PV was developed in a 2T configuration, incorporating light management layers. We developed near-ultraviolet (NUV) and near-infrared (NIR) distributed Bragg reflectors, spectral-selective mirrors, to enhance the performance of transparent photovoltaics without sacrificing transparency, thereby achieving high light utilization efficiency. The transparent photovoltaic devices, consisting of NUV-absorbing perovskite and NIR-absorbing organic photovoltaics, were fabricated by the collaborators. The integrated light-management layers in the 2T-laminated transparent photovoltaic device improved performance, achieving a record-high LUE of 3.65%, a transparency of 30%, and a color rendering index (CRI) of 77. Finally, we demonstrated a prototype window of 60 cm × 60 cm in size as a building-integrated photovoltaic appliance.