Cellulose Nanocrystal Substrates for Recyclable Printed Electronics

Abstract. *Low-cost substrates derived from renewable feedstocks, such as wood, are very attractive for the realization of a sustainable and economically viable printed electronics (PE) industry. Organic solar cells (OSC) are an attractive PE technology to harvest energy from the largest source of renewable energy, the sun. We have fabricated efficient OSC on optically transparent cellulose nanocrystal (CNC) substrates. These OSC reach a power conversion efficiency of 2.7% and can be easily separated and recycled into their major components using low-energy processes at room temperature, opening the door for a truly recyclable solar cell technology.*

Keywords. Cellulose nanocrystals, recyclable electronics, printed electronics, organic electronics, solar cells.

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Introduction. The printed electronics (PE) industry offers a paradigm change in manufacturing and cost compared to the inorganic semiconductor industry because electronic products can be mass-produced from roll-to-roll processes over large areas and on flexible substrates. Availability of cost-effective and environmentally friendly substrates will be vital for the economic viability and future growth of this industry, but simultaneously the environmental footprint of these devices must be minimized. PE devices such as OSC, organic light-emitting diodes (OLED), and organic field-effect transistors (OFET) [1, 2] are made up of thin layers of organic semiconductors and electrodes (typically 10–200 nm thick) that are deposited or transferred onto a thicker substrate (typically 100–200 µm thick) that accounts for most of the total mass of the device. Petroleum-based synthetic polymers such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) among others are widely used as substrates for PE, but they are environmentally less attractive than substrates synthesized from renewable feedstocks such as wood that are easily recyclable or biodegradable.

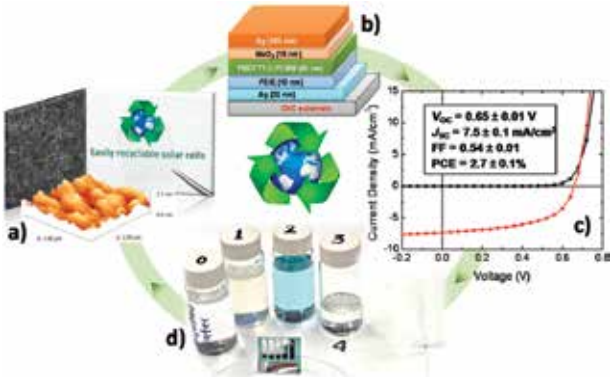

Figure 1. *(a) SEM and AFM image of a CNC film and a picture of CNC film; (b) structure of OSC; (c) J-V characteristics of an OSC on a CNC substrate; (d) recycled solutions and dispersion in vials: Vial #0: distilled water; Vial #1: CNC dispersed in distilled water; Vial #2: photoactive layer in chlorobenzene; Vial #3: chlorobenzene from second wash; #4: MoO_3 and Ag waste on paper filter.*

Cellulose nanomaterials (CN) such as CNCs are high-value materials that can be processed into transparent and flexible films and can be produced in industrial quantities from renewable and sustainable feedstocks [3]. The performance of PE devices fabricated on paper or other CN films so far has been limited. In particular, OSCs fabricated on paper or other CN-based substrates have shown power conversion efficiencies below 1.4% [4,5] due to high surface roughness and porosity of the substrate. Recently, we reported on organic solar cells fabricated on CNC substrates with a power conversion efficiency of 2.7% that are easily recyclable [1].

Methodology. CNC substrates were cast from CNC/glycerol solutions [1]. The surface roughness of CNC samples was characterized by atomic force microscopy (AFM) and the surface morphology by scanning electron microscopy (SEM). OSC devices were fabricated on top of CNC substrates with the geometry shown in Fig. 1b [1]. The Ag and MoO_3 layers were thermally evaporated. PEIE [2] and the PBDTTT-C:PCBM photoactive layer [1] were spin-coated. Current density-voltage (J-V) characteristics of OSCs were measured inside an

N_2-filled glove box in the dark and under illumination from a solar simulator. Recycling of OSCs was demonstrated [1] by immersing a device into distilled water at room temperature. The solution was filtered using a P5 filter paper. The solid waste in the filter was rinsed with chlorobenzene and the waste collected in a vial. Chlorobenzene rinsing was repeated for the second time.

Results. Figure 1a illustrates the high transparency of the CNC substrates using an AFM image displaying a smooth surface with a root-mean-square surface roughness of 1.8 ± 0.6 nm. Figure 1b displays schematics of the OSC geometry. Figure 1c displays the J-V characteristics of an OSC fabricated on a CNC substrate in the dark and under illumination. In the dark, the device displays excellent diode behavior, with low reverse saturation current and large rectification ratio. Under illumination of 95 mW/cm^2 from an AM 1.5G solar simulator, the devices yield a power conversion efficiency (PCE) of 2.7 ± 0.1% averaged over three devices. This represents a significant improvement over previously demonstrated organic solar cells on paper-like or CN-based substrates [4,5], but it is still modest performance compared to devices fabricated on glass or petroleum-based plastics. This can be primarily attributed to the low transmittance of the Ag bottom electrode and is expected to improve significantly with further optimization.

The CNC substrate of an OSC immersed in distilled water quickly swells and dissolves. The redispersed CNC turns into a solid residue on a Petri dish after water has evaporated and can potentially be recovered and recycled. The rest of the layers making up the OSC device turn into a free-standing film or membrane that can be separated by filtration. The organic layers can then be dissolved in an organic solvent and separated from the metal layers by filtering the solution. This allows for the full separation of the solar-cell components (substrate, organic and inorganic materials) at room temperature using a filter paper. Figure 1d displays vials containing the solutions and the final residue on the filter paper to illustrate the final products of this recycling process.

Conclusions. We have demonstrated that CNC substrates can be cast into smooth transparent films with very attractive properties for the realization of PE devices. In particular, we have provided the first demonstration that efficient OSCs can be fabricated on recyclable substrates derived from renewable feedstocks. We have also shown that these OSCs are easily separated into their major components using low-energy processes at room temperature, opening the door for a fully recyclable solar-cell technology. Efficient and easily recyclable polymer solar cells on cellulose nanocrystal substrates could be an ideal technology for sustainable, scalable, and environmentally friendly energy production and could have an overreaching impact on the sustainability of printed electronics. Future work will focus on optimization of the mechanical properties of the CNC substrates and on barrier-coating technology to protect the CNC-based organic solar cells from the environment when they are deployed in the field.

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Cellulose nanocrystals incorporated within the overcoat varnish of the cover!

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