



Editorial: Window Electrodes for Emerging Thin Film Photovoltaics

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Editorial on the Research Topic

Window Electrodes for Emerging Thin Film Photovoltaics

Photovoltaics (PVs) fabricated by printing at low temperature onto flexible substrates are attractive for a broad range of applications in buildings and transportation, where flexibility, color-tuneability, and light-weight are essential requirements. Two emerging PV technologies on the cusp of commercialization are organic PVs and perovskite PVs. CIGS, CdTe, and a-Si solar cells also have potential applications in flexible PVs. It is widely recognized that these classes of PV will only fulfill their full cost advantage and functional advantages over conventional thin film PVs if a suitable transparent, flexible electrode is forthcoming (Lu et al., 2018). Indium tin oxide (ITO) is the most popular transparent conductor material for opto-electronics including solar cells and displays. However, the fragile ceramic nature makes ITO unsuitable for future electronics such as flexible, stretchable, and wearable electronics because it will easily develop cracks under mechanical deformation. Instead, optically thin film or metallic nanowire networks (Sannicolo et al., 2016) of the most electrically conductive metals copper (Cu), silver (Ag), and gold (Au) have shown promising potential, in spite of the oxidation and parasitic absorption problem of Cu and the high material cost problem of Ag and Au. Whilst the chemical, thermal, and electrical stability of transparent electrodes based on these metals presents challenges, it has been shown that thin coating layers can be very efficient in preserving their integrity and properties (Celle et al., 2018). Additionally, low-temperature, high-throughput deposition techniques, such as spatial atomic layer deposition (SALD) (Muñoz-Rojas and MacManus-Driscoll, 2014; Khan et al., 2018), can be used to deposit these protective layers.

The four articles in this Research Topic relate to different aspects of the development of transparent electrodes based on optically thin films of Au (Lucarelli and Brown), Ag (Wang et al.), and Cu (Bellchambers et al.; Pereira and Hatton). In all cases the metal deposition method of choice is thermal evaporation. Given that large area roll-to-roll deposition of metals by vacuum evaporation is an established industrial process for the production of low cost food packaging and insulation foils, it is plausible that this method of metal deposition offers a path to the fabrication of low cost transparent electrodes for PVs.

For films of Cu, Ag, and Au to have high optical transparency and low sheet resistance they must have a thickness $< \sim 10$ nm and a uniform slab-like structure, which is notoriously difficult to achieve on glass and technologically relevant transparent plastic substrates such as polyethylene terephthalate due to the high surface energy of these metals. To address this challenge, various

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approaches have been developed, e.g., with the use of seed layer or introducing metal “doping” to suppress the 3D island growth, leading to smooth metal films (Zhang et al., 2014). In this special issue Wang et al. report a novel nucleation inducing seed layer based on a bilayer of ultra-thin (0.6 nm) Au and polyethyleneimine (PEI) for Ag films, which substantially outperforms either of these nucleation seed layers on their own, reducing the percolation threshold for evaporated Ag films from 6 to 3 nm. Using this approach, in conjunction with a high-refractive index anti-reflection layer, a sheet resistance of $\sim 9 \Omega/\text{sq}$ and a remarkable transmittance of $\sim 93\%$ is demonstrated together with outstanding long-term environmental and mechanical stability. Whilst to date studies reporting hybrid organic-inorganic metal nucleation layers are sparse (Bellchambers et al., 2019). Wang et al. have shown that the benefits can be substantial and in some cases may off-set the disadvantage associated with the extra complexity in fabrication. There is also a very large pallet of material combinations that could be explored and so this is a fertile area for future research.

Perovskite PVs present a particular challenge for metal substrate electrode design because of the chemical incompatibility of most metals toward the iodine-containing compounds used to form narrow bandgap perovskite semiconductors as well as the iodine-containing degradation products (Bastos et al., 2017). For this reason Lucarelli and Brown have used a 10 nm thick film of Au, which offers superior chemical resistance toward oxidation as compared to Ag and Cu. Whilst gold is 80–100 times more costly than Ag, in this context its very low thickness and high stability may justify its use in high performance perovskite PVs since Au is already widely used in the electronics industry. Lucarelli and Brown show that a solution processed SnO_2 layer is particularly effective as a nucleation seed layer for evaporated Au films, and that by depositing the same oxide on top, to match the optical impedance across the metal film, the far-field transparency is greatly improved. SnO_2 is also an inspired choice because it can be interfaced with TiO_2 , which is the most widely used hole-blocking charge extraction layer in perovskite PVs. On plastic substrates these triple layer electrodes offer remarkable stability toward bending through tiny radius of curvature (as small as 1.5 mm) and in PV devices a power conversion efficiencies of 7.6% is achieved.

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Whilst Cu based transparent electrodes are very appealing due to the much lower cost of Cu (1% that of Ag), the susceptibility of Cu to oxidation in air imposes a challenge. To exploit the advantage of low material cost and good electrical conductivity without the oxidation problem (Han et al., 2014). Pereira and Hatton demonstrated a Cu–zinc oxide bilayer electrode supported on flexible polyethylene terephthalate (PET) with a sheet resistance of $11.3 \Omega/\text{sq}$ and average transparency of 84.6% in the wavelength range of 400–800 nm. The Cu film is perforated with a dense array of sub-micron diameter apertures fabricated using polymer-blend lithography, which is found to suppress reflection, particularly for wavelengths $> 550 \text{ nm}$. Compared with ITO, the bilayer electrode showed superior stability toward bending deformation, and organic PVs using this electrode in place of ITO achieved a high power conversion efficiency of 8.7%. With the similar objective to realize high performance Cu based transparent electrodes less prone to oxidation, Bellchambers et al. passivated a 9 nm Cu film electrode with an 0.8 nm aluminum passivation layer, both deposited by simple thermal evaporation. The results showed superior oxidation stability with very little change (3.4%) in sheet resistance after 5,000 h in air because of the segregation of aluminum-copper-oxide to boundaries between the Cu crystallites upon exposure to air, which retards oxidation at those sites in the Cu film most vulnerable to oxidation. These two original studies on new Cu thin film based flexible transparent conductors are expected to accelerate research into low-cost transparent flexible electrodes in numerous emerging optoelectronic devices.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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