Electronic Supplementary Information

Highly efficient, flexible, indium-free perovskite solar cells employing metallic substrates

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Working electrode fabrication

The working electrode was prepared by first manually polishing 150 μm thick titanium foil using an abrasive pad (3M Trizact 3000) to produce a surface with an Ra value of 50-70 nm. This foil was cleaned sequentially by sonication in acetone and isopropyl alcohol, followed by 10 minutes of oxygen plasma. In instances where a compact TiO_2 layer was used, a commercially available precursor solution (Solaronix BL/SC) was spin-coated onto the substrate at 5,000 rpm for 30 seconds, followed by a 2 minute sinter at 550°C. In the cases where a thermally grown TiO_2 layer was used, foil samples were thermally annealed at the same temperature for 10 minutes. In both instances, a colour change from metallic silver to deep blue was observed on the foil surface, indicating the presence of a thin oxide film. Since metallic substrates do not suffer breaking by thermal spalling in the same way glass substrates do, no temperature ramping was required: Samples were simply transferred on and off a pre-heated hotplate.

An insulating scaffold was added next by spin-coating a 4%wt. suspension of Al_2O_3 nanoparticles in 2-propanol onto the foil substrates (2,500rpm for 60 seconds), followed by drying at 150°C for 30 minutes. The organolead halide perovskite layer was produced by spin-coating a 40%wt. solution of methylammonium iodide and lead chloride (3:1 molar ratio) in N,N-dimethylformamide (DMF) onto the substrate under a nitrogen atmosphere. Foils were then transferred to a hotplate and subjected to an annealing procedure involving 10 minutes at 50°C followed by 85 minutes at 100°, and finally 10 minutes at 120°C. Upon the completion of perovskite annealing, iodopentafluorobenzene (IPFB) was spin-coated onto the devices in order to passivate undercoordinated iodine ions on the perovskite surface. A solution of 8% wt. 2,2',7,7'-tetrakis[N,N-di(4-methoxyphenyl)amino]-9,9'-spirobifluorene (spiro-OMeTAD, Borun Chemical) in chlorobenzene was next spin-coated onto the perovskite films. The spiro-OMeTAD solution contained additives including 19mM bis(trifluoromethane)sulfonimide lithium salt (Li-TFSI), 7mM 4-*tert*-butylpyridine (tBP). V_2O_5 powder (99.99%, Sigma-Aldrich) was also added to the spiro-OMeTAD solution in the quantity of 2.5%wt with respect to the spiro-OMeTAD powder. A 10-50 nm thick layer of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) was next deposited onto the hole-transport layer by sequentially spray-coating a 1:2 by volume solution of low-water content PEDOT:PSS (Heraeus GSD1330) and 2-propanol. Multiple spray passes were used to build up a layer of desired thickness, each spray pass was found to deposit a layer approximately 4 nm thick as determined by profilometery. This film was subsequently dried on a hotplate at 55°C so as not to damage the spiro-OMeTAD layer underneath.

Transparent, flexible counter-electrode fabrication

Ex-situ, a flexible transparent, adhesive counter-electrode was prepared by doctor blading a transparent conductive adhesive (TCA) onto a PET film with embedded Ni mesh (Epigem 300S) using a 90 μm height guide. The TCA was prepared by mixing PEDOT:PSS (Agfa EL-P3145) and pressure-sensitive acrylic adhesive (Styccobond F46) 1.3359:1 by weight in a paint shaker. The mixture was then de-gassed under vacuum to remove entrapped air. After casting, the coated film was transferred to a hotplate where it was heated to 60°C for 15 minutes, followed by 120°C for 5 minutes. Upon completion of the heating cycle, the laminate was cut to size and consolidated with the titanium foil electrodes using finger pressure.

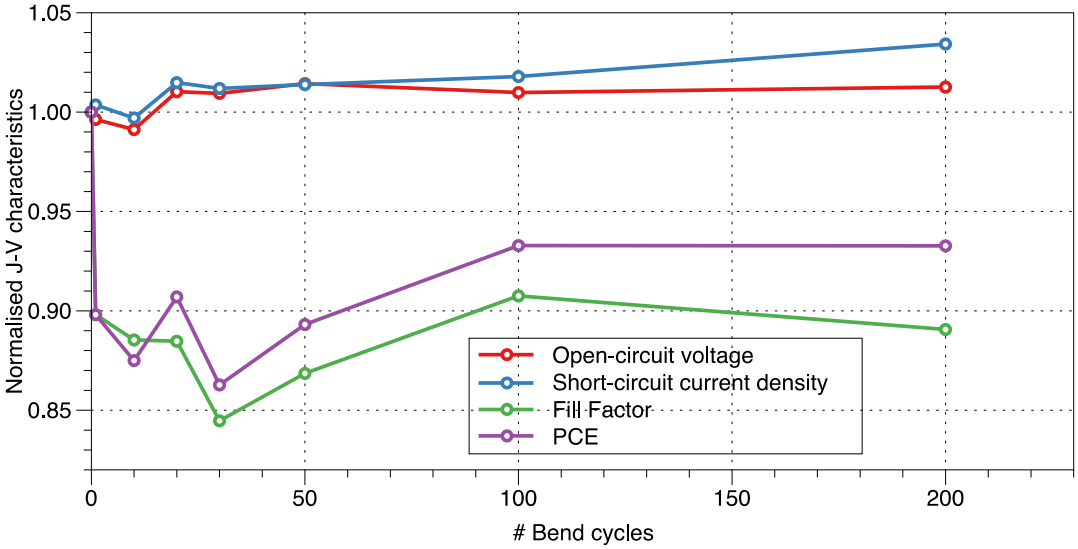

Figure S1. Normalised V_{OC}, J_{SC}, Fill factor and PCE data for a metal-substrate perovskite solar cell undergoing repeated cycles of bend testing around a 5 cm radius

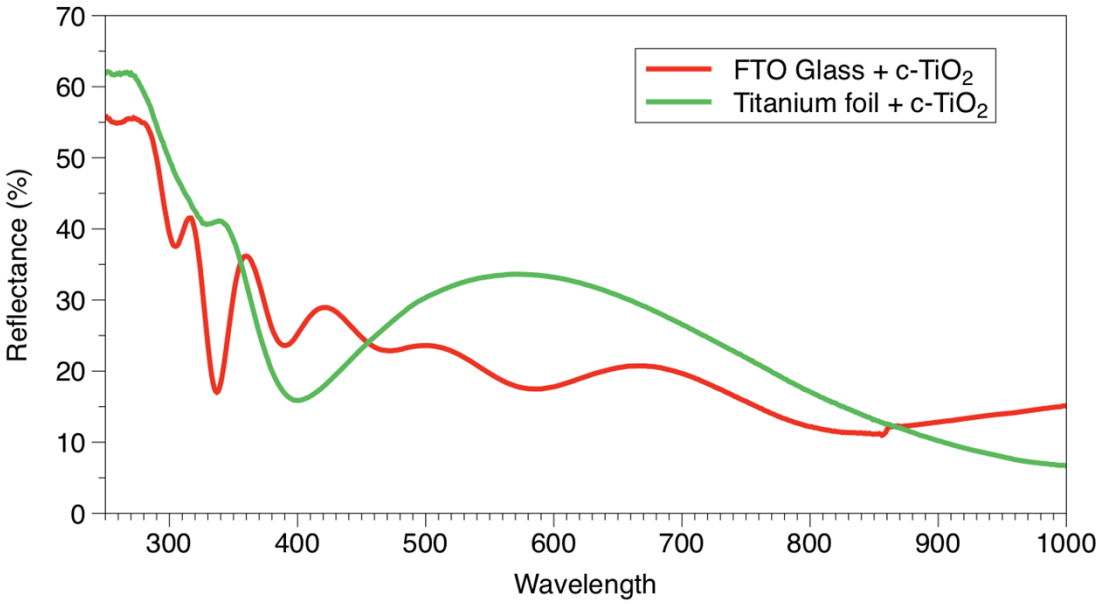

Figure S2. Reflectance spectra for c-TiO2 coated FTO glass (7 Ω sq^{-1}, Pilkington) and smoothed titanium foil

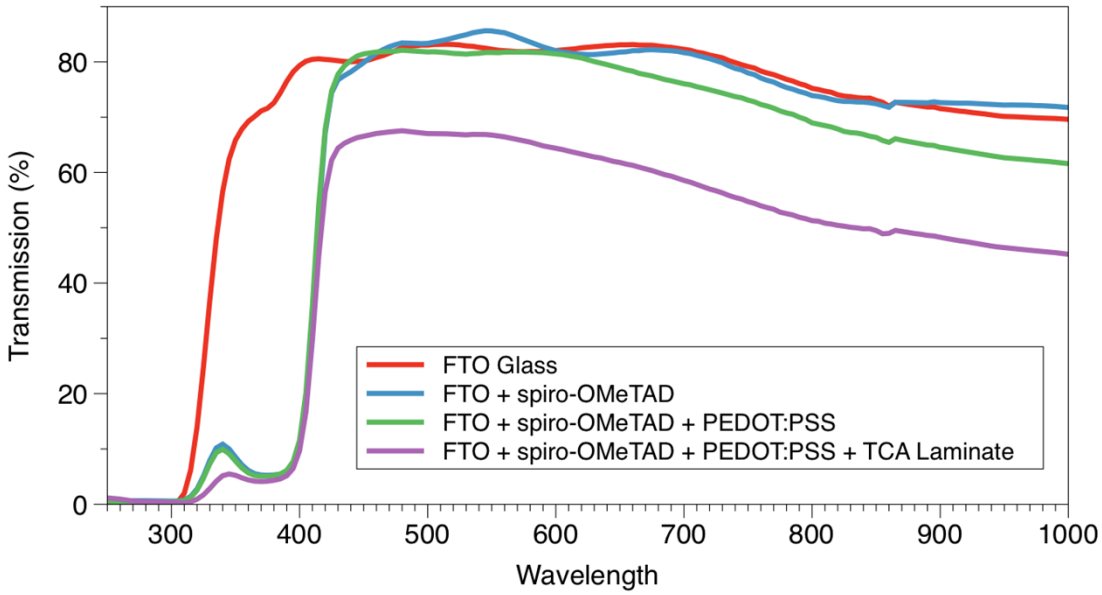

Figure S3. Transmission spectra for FTO coated glass with layers of spiro-OMeTAD, PEDOT:PSS and the TCA laminate applied.

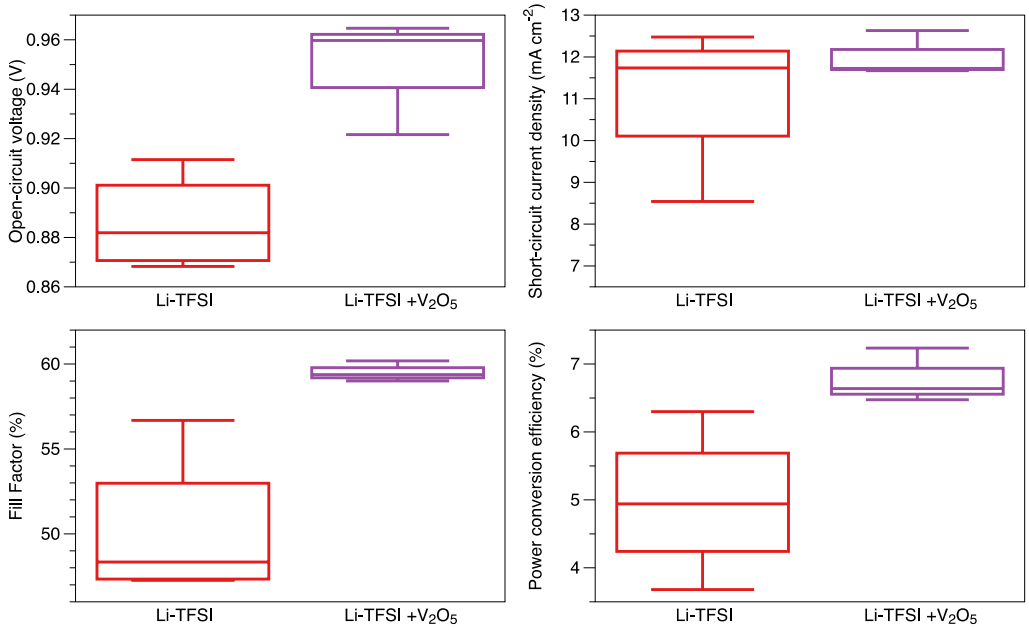

Figure S4. Statistical analysis of J-V data showing titanium-substrate devices with HTM layers doped with Li-TFSI and Li-TFSI with a 2.5wt% addition of V_2O_5 . Devices were tested within 2 hours of HTM deposition. In total, 8 devices were measured.