Large bandgap perovskite solar cells for the application as top cell for Si-PVK tandem devices

Faiazul Haque[1], Kenrick Anderson[1], Angela Chen[1], Benjamin Duck[1], Timothy Jones[1], Noel Duffy[2] and Gregory Wilson[1]

[1] CSIRO Energy, Newcastle Energy Centre, 10 Murray Dwyer Cct, Mayfield West, NSW 2304, Australia.

[2] CSIRO Energy Clayton Laboratories, Clayton, VIC 3168, Australia

Solar photovoltaic technology is steadily growing as a viable renewable energy source to mitigate the ever-increasing energy demand and reduce the adverse effects on the planet done by conventional energy sources. The PV market is currently dominated by single junction crystalline silicon (c-Si) solar cells [1]. By far the maximum efficiency reached by this mature technology has reached 26.7% which is not too far away from the theoretical efficiency limit of 29.4% [2]. Increasing the solar cell efficiency is vital to reduce the levelized cost of electricity (LCOE) hence making PV technology more affordable. As an approach for achieving efficiencies beyond 30%, tandem solar cells are utilised [3]. There are several two-junction tandem architectures reported in the literature such as: Silicon-Perovskite, CIGS-Perovskite, Perovskite-Perovskite etc [4]. Among these, the Silicon-Perovskite tandem technology has shown most promising outcomes in terms of efficiency and commercialization prospects till date and is expected to enter the market very soon [5]. In a Si-Perovskite tandem device a wide bandgap perovskite solar cell is stacked over a low bandgap c-Si cell [6]. This allows the higher energy photons to get absorbed by the high bandgap perovskite top cell and the lower energy photons getting absorbed by the low bandgap silicon bottom cell ensuring superior utilisation of the solar spectrum compared to a single junction device. A two-junction tandem solar cell can theoretically yield efficiencies up to ≈46% [7].

The options of bandgap tunability and comparatively cheaper and easier fabrication techniques offered by the perovskite (PVK) solar cell technology have proven to be an ideal companion as a top cell on top of the Si bottom cell. By far the maximum efficiency reported from a Si-PVK tandem is 30% in a laboratory scale which is a promising figure indeed to break the '30% psychological barrier' already [8]. Now the next big thing will be the commercialisation of this high efficiency technology so that solar PV can also increase its lead as a viable renewable resource to reduce carbon emissions further.

CSIRO is also working towards Si-PVK tandem technology to produce on a large scale for commercialization in the near future. As a first step towards this target, wide bandgap (1.68 eV) semitransparent perovskite top cell optimisation was carried out in a laboratory scale which can be later translated towards the tandem architecture with a Si bottom cell. An inverted p-i-n architecture was chosen as the perovskite cell architecture in this work as they have proven to be more compatible and efficient in the tandem architecture since their n-i-p counterparts have shown to have more parasitic absorption losses coming from the hole transport layer facing the sun when employed as a top cell stack in the tandem cell [9]. Triple-cation lead mixed halide [$Cs_y(MA_xFA_{1-x})_{1-y}Pb(I_zBr_{1-z})_3$] perovskite layer was used as the active layer. 2PACz and C60 were selected as the hole transport and electron transport layers, respectively. The schematic of the device structure is shown in Figure 1. Patterned glass/ITO were used as substrates. The HTL and perovskite layer was deposited by spin coating. The ETL was thermally evaporated over the glass/ITO/HTL/Perovskite stack. Finally, the devices were completed by evaporation of Ag metal electrodes. Figure 2 shows the top view of a completed lab scale high bandgap perovskite solar cell fabricated in the CSIRO lab with both small and large area devices. Several optimisations were

carried out on the HTL deposition parameters such as concentration of 2PACz, spin coating speed, rest time variation etc. Different composition of the perovskite active layer was also investigated by tuning the composition of the perovskite and maintaining the desired bandgap. Finally, the optimised device stack demonstrated an impressive efficiency of 17.63% and 15.90% for small area (0.13 cm^2) and large area (1 cm^2) cells. The repeatability of the optimised fabrication parameters was also confirmed for more than 10 devices from a single batch showing similar output which proves the superior reproducibility of the fabricated devices. The best J-V curves for small and large area devices are shown in Figure 3. The morphology of the optimised perovskite layers obtained by SEM are shown in Figure 4. The spectral response and bandgap of the fabricated perovskite layers from EQE (Figure 5) aligns well with the literature proving the suitability of the optimised thin films for application in the tandem devices.

For stability measurements, the cells were sealed and kept inside Memmert climatic test chamber. The temperature was cycled between -40 °C ± 2 °C and 85 °C ± 2 °C within 8 hours consisting of a single cycle. The normalised PCE of the cells retained >80% of the initial value after 50 cycles (400 hours). Figure 6 shows the thermal cycling profile of temperature and relative humidity for 1 cycle and the J-V curves taken at the beginning, 200 hours and 400 hours, respectively.

This work demonstrates CSIRO's approach towards the commercialisation of Si-perovskite tandem cell research. This optimisation is the primary step for developing a reproducible recipe of the wide bandgap perovskite top cell fabrication which will be transferred in the tandem architecture with a silicon bottom cell. Further optimisation will be required in a monolithic configuration and several other layers arising when employed in a tandem stack such as recombination layer, buffer layer etc. There is also room for optimisation in the silicon bottom cell structure and doping levels to ensure maximum current matching and incident photon utilisation for maximising the efficiency of the overall stack.

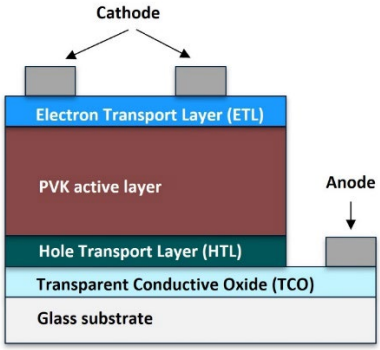

Figure 1: Schematic of the device structure

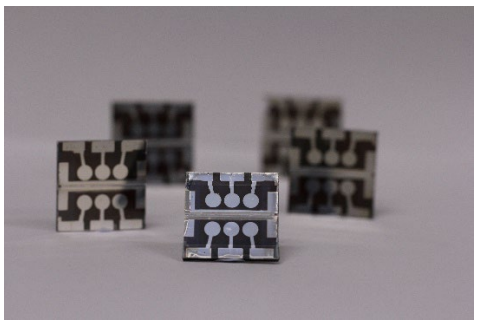

Figure 2: Top view of the fabricated high bandgap perovskite devices in the lab

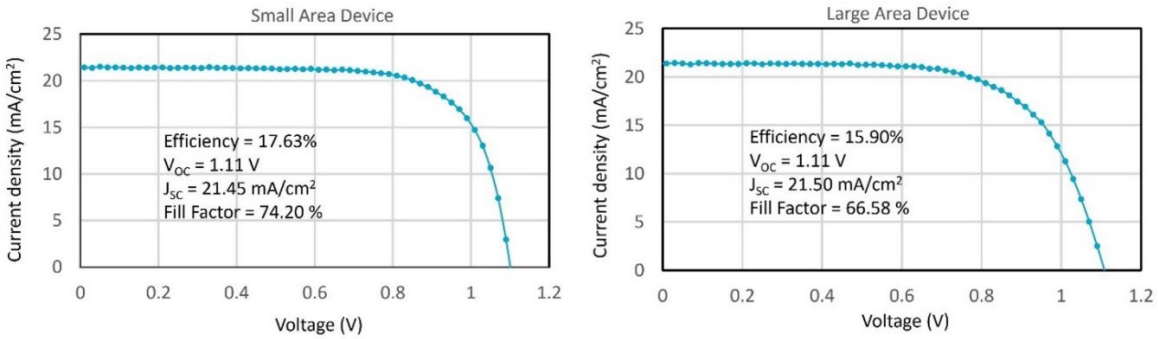

Figure 3: J-V curves for small and large area devices

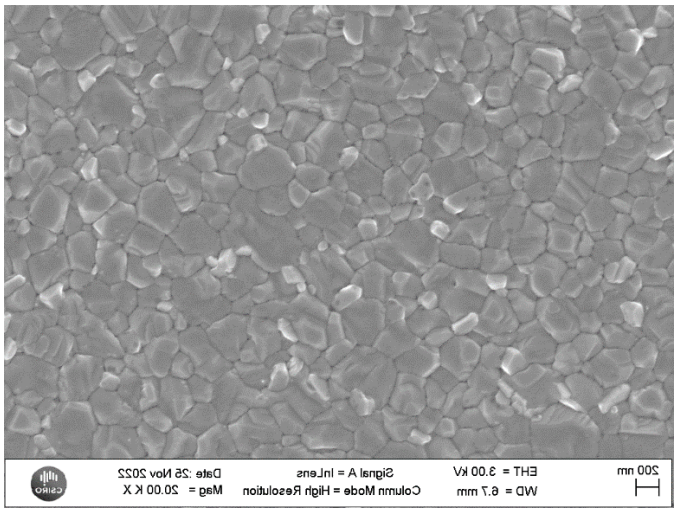

Figure 4: Morphology of the active perovskite layer from SEM

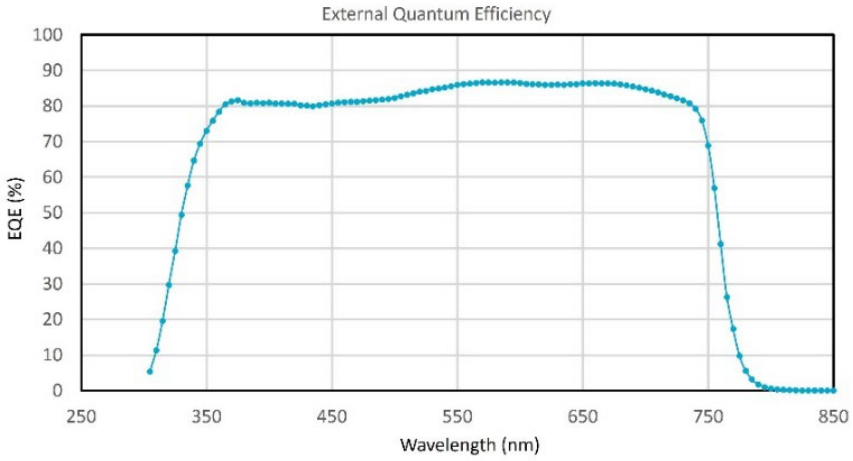

Figure 5: EQE spectra of the fabricated perovskite devices

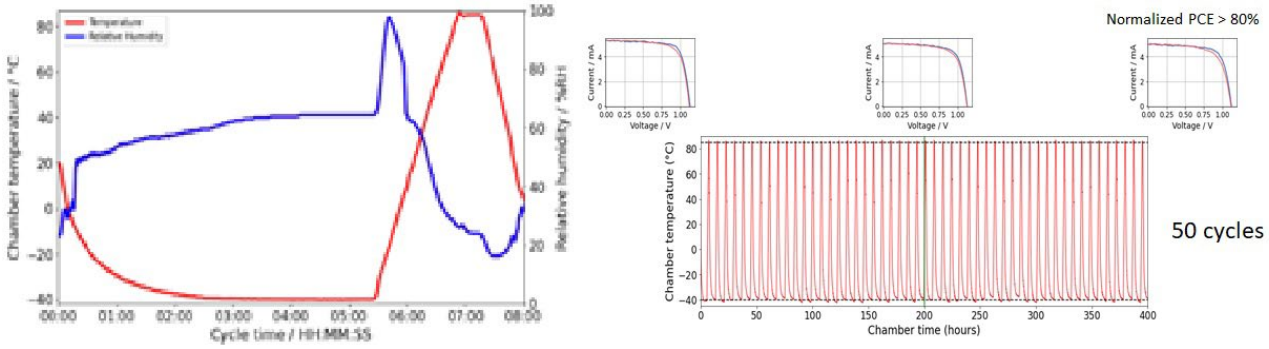

Figure 6: Thermal cycling test profile and J-V curves taken at different times

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