

## Monolithic integration of two-terminal perovskite/Cl(G)S tandem cell and interface optimization

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The PERCISTAND project is a European collaboration whose aim is to reach 30% power conversion efficiency (PCE) with perovskite/Cl(G)S tandem solar cells within three years, in both two- and four-terminal architecture.

In the context of two-terminal (2T) perovskite/Cl(G)S tandem cells, a junction can be formed between n-type IZO on top of the Cl(G)S subcell and p-type NiO at the bottom of the perovskite subcell. To avoid damaging the Cl(G)S subcell, the post-annealing treatment of any subsequent layer should not exceed 150°C. It should also be noted that the root mean square (RMS) roughness on the surface of as-prepared Cl(G)S layers can reach values as high as 60 nm.

Herein, for the top cell, we present several strategies to prepare suitable NiO layers (sputtering, evaporation, sol-gel, nanoparticles) and subsequent perovskite top cells. Furthermore, by design atomic layer deposition (ALD) is a method which we tested and which yields homogeneous thin layers on rough substrates. ALD-prepared NiO is often post-annealed at 300°C but Jost et al. demonstrated the possibility to reach a 21.6% PCE with voltage additivity for 2T perovskite/Cl(G)S tandem cells using a pristine ALD layer<sup>[1]</sup>. The sputtering method can also yield homogeneous NiO films can also be deposited on rough substrates. To optimize low-temperature development of NiO by ALD sputtering, the electrical and chemical properties of various films prepared under different conditions has been investigated by XPS and KPFM.

For the bottom cell, chemical etching can be used directly on rough Cl(G)S layers in order to deposit subsequent homogeneous films by wet process. In 2011, by using a bromide solution formulation of HBr/Br<sub>2</sub>/H<sub>2</sub>O Bouttemy et al. were able to heavily reduce the RMS roughness of Cl(G)S layers from 200 nm to 50 nm<sup>[2]</sup>. In 2020, Kamikawa et al. reported a PCE of 19% using a chemically etched absorber layer whose roughness was reduced to tens of nanometers<sup>[3]</sup>. Here we report the fabrication of Cl(G)S solar cells based on the method developed by Bouttemy et al. to obtain RMS roughness as low as 18 nm on 4 cm<sup>2</sup> substrates. By optimizing the process, we were able to minimize the thickness of the sacrificial layer and obtain almost identical IV parameters for solar cells derived from both flattened and non-flattened absorber layers.

<sup>[1]</sup>: M. Jost et al., 21.6%-Efficient Monolithic Perovskite/Cu(In,Ga)Se<sub>2</sub> Tandem Solar Cells with Thin Conformal Hole Transport Layers for Integration on Rough Bottom Cell Surfaces, *ACS Energy Lett* 4 (2019) 583–590

<sup>[2]</sup>: M. Bouttemy et al., Thinning of Cl(G)S solar cells: Part I: Chemical processing in acidic bromine solutions, *Thin Solid Films* 519 (2011) 7207–7211

<sup>[3]</sup>: Y. Kamikawa et al., Efficient narrow bandgap Cu(In,Ga)Se<sub>2</sub> solar cells with flat surface, *ACS Appl. Mater. Interfaces* 12, 40 (2020) 45485–45492