Rethinking chalcopyrite solar cells architecture for solar fuel production

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Latest developments of the chalcopyrite CIGSSe based photoelectrodes make it one of the most promising choices for photoelectrochemistry applications such as CO2 reduction as recent efforts showcased promising Faradic efficiencies at low bias potentials. [Hu et al. Journal of Catalysis, 2020 ; Robert et al. Nature Communications, 2020]

However, CIGSSe technology has been developed mainly for photovoltaic application. Consequently, the prevalent device configuration encounters significant drawbacks when used in photoelectrochemical cell (PEC) systems.

Firstly the opaque back contact implies the device to work under front side illumination -restricting the choice of compatible catalysts and grafting surfaces to non-absorbing materials; secondly the AZO upper layer experiences limited resilience to PEC operating conditions (i.e. aqueous solutions, pH…). Finally, CIGSe's bandgap is too small to perform water-splitting and CO2 reduction - requiring to employ an additional power source.

We thus embarked on the development of chalcopyrite solar cells with a revised architecture: the wider-bandgap sulfide CIGS is grown on a transparent conductive oxide (TCO), so the device can be illuminated through this transparent back contact and a much larger varieties of upper layers and catalysts can be employed.

Significant efforts have already explored CIGSe based solar cells on TCO (usually ITO) for tandem or bifacial application. But the formation of secondary phases during CIGSe films growth compromises the ITO/CIGSe interface, leading to substantial photocurrent loss.

Here, we worked on both the materials and the device preparation to obtain solar cells based on an ultrathin for which no indication of TBC/absorber performance bottleneck is found.The resulting solar cells have a photovoltaic efficiency of 11% under rear-illumination, close to the 12.6% obtained with the same material and a standard cell architecture (i.e operating under front illumination).

The addition to these solar cells of cobalt quaterpyridine molecular catalyst immobilized on a multi-walled carbon nanotube layer results in PEC devices. Those hybrid CIGS photocathodes achieve unprecedented photocurrent densities for CO2 reduction (several times the state of the art). Catalytic performances will be described in this presentation.