



Photoelectrochemical water splitting: an idea heading towards obsolescence?

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The production of hydrogen from water and sunlight is a way to address the intermittency in renewable energy production, while simultaneously generating a versatile fuel and a valuable chemical feedstock. Photoelectrochemical water splitting is one possible approach to accomplish this that has been researched since the early seventies. It has for a long time held the promise of having the potential to become the best, cheapest, and most efficient way to convert solar energy into chemical energy in the form of hydrogen, but in this paper, I argue that the time window where this could have happened has now come to an end. With the rapid development of both PV-technology and earth-abundant electrocatalysis, it will be tremendously difficult, even in the best-case scenario, for a classical photoelectrochemical water splitting device to compete with what PV-driven electrolyzers can already do today. This is an insight that should influence the future of solar fuel research.

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Broader context

In a time where our sources of oil are running dry, where soot from burning coal is poisoning the air, and where keeping anthropogenic climate change at bay have reached the political agenda at the highest level, the demand for a transition into a sustainable energy system has never been greater. One conceptual solution to the problem is what is known as the hydrogen economy, where hydrogen is utilized as an energy carrier, a versatile fuel, and a means to balance the inherent intermittency from renewable energy production. To come true, this vision requires sustainable hydrogen production. Given that the sun is the most abundant energy source available, a straightforward question is how to most cheaply and efficiently capture that energy and store it in hydrogen. That is the question being discussed here.

Sustainable production of hydrogen from solar energy and water is a matter of separation, transport, and transfer of photogenerated charge carriers. This requires a photoabsorber, a pair of catalysts, and current transport between them. The straightforward solution is to use a pair of electrocatalysts for electrolysis of water, a process known since 1800, with a solar cell providing the electricity, just like in the toys sold at science museums (Fig. 1a). Photoelectrochemical water splitting (PEC-WS) aims at accomplishing this cheaper and more efficiently by monolithically integrating the photoabsorber with the catalysts, like in Fig. 1b or in, for example, Nocera's artificial leaf.¹ The core of this perspective is that PEC-WS no longer is a good idea, but is heading towards obsolescence due to the remarkable development of silicon PV-technology.

PEC-WS research started in the seventies with the exploration of new photoelectrode materials. From a historical perspective, this made sense. PV-technology was expensive and required advanced manufacturing processes, whereas metal oxide photoelectrodes could be deposited by simple methods and required fewer

solid-state layers. The requirements of an ideal material are demanding and involve a sufficiently high band gap, band edges straddling the redox potential of water, good transport properties, high catalytic activity, and stability in the electrolyte, as well as abundance, non-toxicity, and cheapness. This is a difficult set of constraints, but the band gap requirement alone puts the single-junction PEC-WS approach at a disadvantage.

Thermodynamically, water splitting requires 1.229 V and with overpotential losses, an electrochemical bias of at least 1.5 to 2.0 V is required (Fig. 1c). For a one-cell single-junction PEC-device, this translates into a band gap of 1.9–2.3 eV or more.² A band gap of 2.3 eV gives a maximum current density corresponding to 10% solar-to-hydrogen (STH) efficiency, whereas a PV-electrolyser based on series interconnected silicon PV-cells and a Ni-catalyst can reach over 15%.³ 1.9–2.3 eV is simply too far away from the 1.35 eV for optimal power conversion for a single junction cell given the sun's spectral distribution.⁴ This puts PEC-WS at a disadvantage, but the real game changer is the relation between efficiency, module price, and balancing of system (BOS) costs. The record price for PV-electricity is now below 0.03 \$ per kW h and is still falling,⁵ thus taking silicon PV towards providing some of the cheapest electricity on the planet.⁶

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opportunity there was, whereas progress in PV-electrolysis has made even the most optimistic targets obsolete. I am claiming this, not as a sceptical outsider, but as a previous enthusiast with years of research in this endeavour, who thinks it is time to accept the inevitable and move on towards other intellectual challenges and more promising enterprises. The PEC-WS research conducted over the last few decades has resulted in much insight into, for example: heterogeneous catalysis, reaction mechanisms, charge transfer processes, semiconductor photo-physics, *etc.* PEC research is still worth doing, but it would benefit from shifting the focus from the water splitting narrative towards the specific phenomena investigated, which could be interesting in their own right.

There is a chance that I am wrong, and I am sure not everyone will agree, but the important insight is that if PEC-WS is to have even a shadow of a chance, it will need to operate very close to its fundamental thermodynamic limits. There is simply no room for compromises – whatsoever. For the ones still clinging to the PEC-narrative, I would like to challenge you with what may be the greatest intellectual challenge in the field of solar fuels, the one of demonstrating how a large-scale PEC-WS installation could possibly give a significant advantage over a PV-electrolyser. I, and the rest of the world, are eager to listen.

Conflicts of interest

There are no conflicts to declare.

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