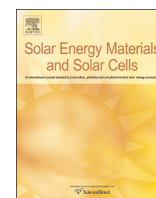




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A theoretical analysis of optical absorption limits and performance of tandem devices and series interconnected architectures for solar hydrogen production

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ABSTRACT

Photo-driven catalytic (PDC) water splitting, using either photoelectrochemical cells (PEC-cells), PV-electrolyzers, or some hybrid system in-between, has attracted a lot of attention. In single-cell device architectures for solar hydrogen production, based on single band gap photoabsorbers, there is a fundamental efficiency problem originating from the energy distribution of the solar spectrum and the thermodynamic and kinetic requirements for splitting water. The minimum band gap for a single-junction device in order to withhold unbiased overall water splitting is considered to be at least 2.0 eV. This is far from the 1.35 eV which is the optimal band gap of a semiconductor for maximum power conversion of light in the solar spectrum. This discrepancy has been termed as the solar spectrum mismatch problem (the SSM-problem). The standard solution to this problem is to construct tandem devices, whereas an alternative is to interconnect several one band gap cells in series, side by side. Both approaches enable the use of low energy photons in the solar spectrum while still providing a sufficiently high photopotential for driving the full reaction, without seriously compromising with the area efficiency.

In this paper, the tandem and serial architectures for handling the SSM-problem are analyzed and compared. The analysis is focused towards differences in the limits of optical absorption, the optimal number of optical absorbers, and their corresponding band gaps. Taking losses due to charge carrier separation and catalysis into account, the maximum STH-efficiency for a series interconnected solar splitting device was found to be 24.6%, compared to 32.0% for an optimum tandem device at 1 Sun (air mass 1.5, 1000 W m^{-2}). This can be compared with the maximum efficiency of 18.0% for an ideal single band gap photoabsorber in single junction device. The analysis shows that the maximum STH efficiency limits for series interconnected architectures for unassisted solar water splitting are not particularly far behind the more commonly studied tandem devices. They could then be an interesting alternative given the simplicity and versatility of series interconnected device architectures. The analysis also compares how tandem devices and series interconnected devices can differ in terms of charge carrier separation, charge carrier transport, catalysis, overall efficiency, device architecture, and expected cost.

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1. Introduction

In a sustainable future, hydrogen has the potential to be a key energy carrier. That future is known as the hydrogen economy [1–3], and to realize this, sustainable means of hydrogen production must be further developed. That is in contrast to the present, non-sustainable, situation where approximately 96% of the global hydrogen production is based on steam reforming of fossil fuels [4]. Examples of possible routes towards sustainable hydrogen include fermentation of biomass

[5–7], thermochemical processes [8–10], and photobiological water splitting [11–13]. The focus in this article is, however, on inorganic photo-driven catalytic hydrogen production (IPDC hydrogen production), i.e. photoelectrochemical cells (PEC-cells) and PV-electrolyzers. In particular, different device strategies for combining a high light harvesting in the solar spectrum, leading to high photocurrents, with a sufficient photopotential to drive the full reaction are explored and analyzed.

The conceptually simplest device for solar hydrogen production is probably a one band gap, single-junction, monolithic PEC-device. Ideally, that is a single photoabsorber which when illuminated and immersed in water provides the electrochemical

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driving force for both the oxidation and the reduction of water. The investigations and development of such devices have been pursued since the seventies, and the starting point of the historical narrative of the PEC-cell is often taken as the 1972 Nature paper by Fujishima and Honda [14], demonstrating photo-driven water oxidation using TiO_2 .

The single-junction devices have a fundamental efficiency problem. The thermodynamics and kinetics of the water splitting reaction require the band gap of the photoabsorber to be at least 2 eV [15–17], although higher limiting values also have been suggested [18–20]. That is considerably higher than the 1.35 eV that would be optimal for maximum power conversion of the solar light while using a one band gap device [21]. The spectral distribution of the solar spectrum, given by the surface temperature of the sun, is hard to change, as is the thermodynamics of the reduction and oxidation of water. This incompatibility is here termed the solar spectrum mismatch problem (the SSM-problem), and is a fundamental problem for single-junction devices. Such a device will consequently face difficulties in comparison to alternative routes towards sustainable hydrogen in terms of efficiency, and thus also in the total cost per produced unit of hydrogen.

An often proposed solution to the SSM-problem is to construct tandem devices, where photoabsorbers with different band gaps are stacked on top of each other. The tandem cell architecture can increase the photopotential, i.e. the difference in the chemical potentials of the photogenerated charge carriers, up to what is required for the full reaction. A larger part of the energy in the solar spectrum can thus also be utilized compared to a device based on a single-junction photoabsorber.

An alternative to tandem devices for solving the SSM-problem is to interconnect several single-junction cells in series, side by side. By interconnecting cells in series, the photopotential can be increased up to what is required for driving the full reaction, without seriously compromising with the area efficiency of the corresponding single band gap cell. Semiconductor materials with optimal band gap for solar energy harvesting and promising charge carrier properties can then be utilized in devices for hydrogen production.

The knowledge and function of serial interconnected solar cells and their use in solar-to-electricity conversion is well known. This is, however, knowledge that to a very small extent has been used in the context of solar hydrogen production where two chemical reactions have to be performed at the terminating surfaces. The optimum efficiency for delivering electrons and holes to match two reactions at the surface with certain overpotential is not the same as the conditions for obtaining the maximum overall power from a single junction or tandem device. Series interconnected cells turns out to be a promising alternative to the commonly used tandem architecture. We have previously constructed devices based on series interconnected CIGS modules which were demonstrated to have a STH-efficiency of 10% [22]. During the last two years we have presented results on solar water splitting using series interconnected cells and advocated this idea, which previously has been more or less totally absent from the main stream water splitting community. This represents a so far rather neglected route within the solar fuel field, even though different groups now start to pick it up and utilize these ideas. Our previous [22] study has for example been quoted with respect to this issue both in PNAS and Science papers during 2014 [23,24]. There is, however, still a need for deriving the basic framework for the maximum efficiencies that can be expected for series interconnected cells in the context of solar hydrogen production.

In this paper, these two approaches for handling the SSM-problem, i.e. tandem devices and single-junction cells interconnected in series, are analyzed and compared with single band gap photoabsorbers in

the conventional single junction PEC cell. One of the largest differences between the concepts lies in the optical absorption limit, which is analyzed in some detail. Also the difference and similarities in the charge carrier separation, charge carrier transport, catalysis, efficiency, stability, and possible device architectures are analyzed.

2. Discussion

2.1. Absorption in single-cell, one band gap devices

The splitting of water into molecular hydrogen and molecular oxygen using solar radiation can conceptually be divided into four fundamental processes: charge carrier generation, charge carrier separation, charge carrier transport, and charge carrier transfer at the electrolyte interface [25]. An external quantum efficiency for the reaction, EQE, can then be defined as in Eq. (1) where ϕ_{gen} is the yield of charge carrier generation or light harvesting efficiency, λ is the wavelength, ϕ_{sep} is the charge separation efficiency, ϕ_{trans} is the charge transport efficiency, and ϕ_{cat} is the quantum efficiency of the catalytic charge transfer to the desired redox species in the electrolyte.

$$\text{EQE}(\lambda) = \phi_{\text{gen}}(\lambda)\phi_{\text{sep}}(\lambda)\phi_{\text{trans}}\phi_{\text{cat}} \quad (1)$$

The Gibbs free energy for the water splitting reaction in Eq. (2) is 237.18 kJ/mol producing H_2 , corresponding to a thermodynamic reversible potential of 1.229 V.



In order to drive this reaction in practice, an additional potential difference is needed between the anode and the cathode, due to losses associated with charge carrier separation (η_{sep}), transport (η_{trans}), and catalysis (η_{cat}). The useful energy difference between the photogenerated electron and the hole in a semiconductor is the difference between the quasi Fermi levels for the electrons and holes under illumination at the different electrode surfaces, rather than the difference in energy between the band edges within the photoabsorber. This could be seen as the energy penalty for withholding the electric field separating the electrons from the holes. A formulation of this loss, (η_{sep}), was derived by Shockley and Queisser [26] and by neglecting higher order terms can be written as Eq. (3) [20].

$$V_{\text{oc}} = E_g - \eta_{\text{sep}} = E_g - k_B T \ln \left[\frac{8\pi(k_B T)^2 n^2 E_g}{c^2 h^3 j_{\text{gen}}} \alpha L \phi_{\text{rec}} \right] \quad (3)$$

where E_g is the band gap, k_B is Boltzmann's constant, T is the temperature, h is Planck's constant, c is the speed of light in vacuum, n is the refractive index, j_{gen} is the rate of photon absorption in the AM1.5 spectra, ϕ_{rec} is the ratio between the nonradiative and radiative recombination rates, α is the absorption coefficient, and L is the minority carrier diffusion length and should be replaced with the material thickness, d , if $d < L$. Several parameters are material dependent and the loss is then naturally dependent on the choice of photoabsorber and the purity of the material. Typical experimental values for state-of-the-art materials in solar cells are as follows: 0.36 eV in silicon, 0.3 eV in GaAs, 0.4 eV in InP, 0.45 eV in CIGS, 0.6 eV in CdTe, and 0.61 eV in amorphous silicon, and correlate reasonably well with the theoretical assumptions in Eq. (3). To reduce the dimensionality in our data we have used $\eta_{\text{sep}} = 0.4$ eV in our subsequent analysis as a prototypical limiting value. This is in the right order for most PV-materials [21,27] and accounts for the difference between the band gap energy and the open circuit potential. The value can, without loss of generality of the analysis, be replaced with the

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