



Porous multi-junction thin-film silicon solar cells for scalable solar water splitting

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ABSTRACT

Monolithic solar water splitting devices implemented in an integrated design approach, i.e. submerged in the electrolyte, pose a significant limitation when it comes to up-scaling. The ion transport distances around the monolith are long and consequently, the ionic Ohmic losses become high. This fact turns out to be a bottleneck for reaching high device efficiency and maintaining optimum performance upon up-scaling. In this paper, we propose a new device design for integrated monolithic solar water splitting based on porous multi-junction silicon solar cells. Simulation results highlight that porous monoliths can benefit from lower ionic Ohmic losses compared to dense monoliths for various pore geometries and monolith thicknesses. In particular, we show how micrometer scale pore dimensions could greatly reduce Ohmic losses, thereby minimizing overpotentials. A square array of holes with a diameter of 20 μm and a period of 100 μm was fabricated on single-junction and multi-junction amorphous and microcrystalline silicon solar cells. A small impact on the open circuit voltage (V_{oc}) and short circuit current density (J_{sc}) was obtained, with porous triple junction solar cells reaching V_{oc} values up to 1.98 V. A novel device design is proposed based on porous triple-junction silicon-based solar cells.

1. Introduction

Electrification offers a great potential to enable the de-carbonisation of energy [1]. According to several studies, a sustainable energy system based on 100% renewable energy sources, of which the majority is wind and solar, should be possible [2,3]. However, the intermittent nature of wind and solar is a bottleneck for large-scale grid implementation. In order to balance generation and demand, the storage of produced energy is imperative. The storage of renewable electricity in the form of chemical bonds, such as hydrogen (H_2), could fill in the gap and empower a sustainable energy system [4]. Other than storage, green hydrogen could play an important role in such a renewable energy system for the de-carbonisation of transportation, building heating and as fuel for industrial use.

Among the many different techniques which can achieve the conversion of water and sunlight into hydrogen (e.g. PV-electrolysis, thermal conversion and molecular artificial photosynthesis), photoelectrochemical (PEC) water splitting is a promising approach [5–8]. Apart from the properties and performance of the involved materials in

a PEC device (photoabsorbers with optimal band gaps, catalysts, protective coatings, etc.) the cell design is a key element not only for enabling high solar to hydrogen efficiency (STHE, i.e. the ratio of the available chemical energy in the generated hydrogen to the incident solar energy), but also for maintaining those high values when up-scaling for large-scale implementation.

The majority of lab scale PEC demonstrators employ the wired configuration, shown in Fig. 1a. With this design, Khaselev et al. have demonstrated a device reaching an STHE of 16.4% [9]. However, their approach includes expensive III-V materials which are too costly for terrestrial applications. In a cost-effective approach based on triple junction silicon solar cells, the same group showed a device reaching 7.8% [9], while more recently Urbain et al. reported a device reaching an STHE of 6.8% [10]. In order to reduce cost, the wireless monolithic design (Fig. 1b) has been proposed. With this design, Reece et al. reported a 2.5% efficient PEC device [11]. They attributed the lower STHE value compared to their wired reference device, which reached an STHE of 4.7%, to the high Ohmic losses due to long ion transport distances around the monolith. In fact, the ionic Ohmic losses are

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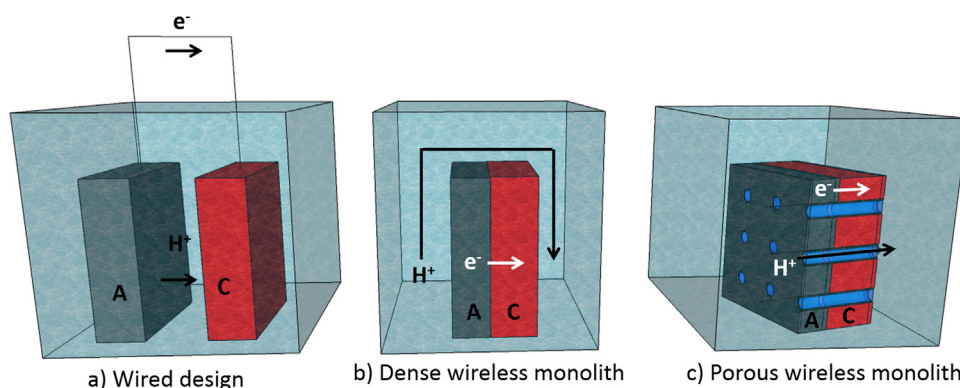


Fig. 1. Photoelectrochemical cell configurations for monolithic integrated solar water splitting: (Photo)anode (A) and (Photo)cathode (C) in a) a wired configuration where the ionic transport distance is defined by the distance between the two photoelectrodes, usually in the cm range, b) the monolithic wireless approach where the ionic transport distance is defined by the thickness and surface area of the monolith and c) porous monolithic wireless configuration where through plane pores act as ionic shortcuts and the ionic transport distance can reach a few tens of μm .

higher in a wireless than a wired design (electronic conductivities are much higher than the ionic conductivities), thus limiting the efficiencies [12]. However, this approach offers an alternative to satisfy the efficiency/cost trade-off [4]. Towards this direction of integrated monolithic devices, the idea of porosity (Fig. 1c) for solar water splitting has been mentioned conceptually [7,12], has been demonstrated in simplified monoliths [13], while theoretical works have touched upon the concept of ionic shortcuts [14,15]. In particular, as discussed by Haussener et al. [14], for monolithic PEC configurations, small electrode lengths and thin separation distances lead to reduced Ohmic losses because of short ion transport distances. For an operation sustained by water vapor, a monolith width of less than $300\ \mu\text{m}$ is proposed for reducing the overpotential to values lower than $100\ \text{mV}$, even for a thickness of $1\ \mu\text{m}$ [15]. According to such design guidelines, through plane pores incorporated on the monolith could act as ionic shortcuts, minimizing Ohmic losses. To further investigate this idea, simplified perforated monoliths were fabricated experimentally, and their Ohmic losses were evaluated. As shown by Bosserez et al. [13], the Ohmic losses were kept at acceptable levels (below $100\ \text{mV}$ of overpotential) provided a pore spacing less than $1\ \text{mm}$ and electrolytes at extreme pH values.

In this paper, we propose a novel device design based on porous multi-junction thin-film silicon solar cells. In such a design, the multi-junction solar cells could provide the required high voltage values for the water splitting operation while micron-scale porosity would act as a shortcut for ions, minimizing their transport distances and thus, the respective Ohmic losses. Therefore, the achieved STHE values could be maintained upon up-scaling for cost-reduction. Our intention is to provide a new design approach, based on the concept of micron-scale porosity, so as to trigger discussions and activities within the scientific community. Such a paradigm shift in the device design of monolithic PEC cells could hopefully help to further develop new designs deviating from more conventional concepts.

In the following sections, we will present our simulation results on porous monoliths so as to highlight the potential of this approach. Furthermore, we will present the experimental fabrication of micron-scale pores on single-junction and multi-junction thin-film silicon solar cells and we will discuss the impact of introducing porosity on their performance. Finally, we will discuss the perspectives of this approach and present our envisioned device design.

2. Material and methods

In order to investigate the potential of the concept of porous PEC monoliths, 2D electrochemical (EC) simulations were performed using the Tertiary Current Distribution electrochemistry module of COMSOL Multiphysics [16]. In our simulations, the system was considered to be periodic in the horizontal direction with monoliths of various periodicity (P), pore width (w) and thickness (t) operating at electrolytes with different pH , as shown in Fig. 2. The reaction kinetics at the anode and

cathode sides were considered to follow the Butler-Volmer equation. The pore sidewalls were considered to be perfectly insulated and do not act as reaction sites. In order to focus only on the effect of the geometrical parameters on the monolith's performance, we restricted the simulations on state-of-the-art high performing reaction kinetics considering Pt electrode parameters for the cathode and RuO_2 parameters for the anode.

The current flows in the EC system through ion transport in the electrolyte were modelled with $J_i = F \sum_{i=1}^n z_i (-D_i \nabla c_i - z_i \mu_{m,i} F c_i \nabla \phi_i)$, where J_i is the electrolyte current, F is the Faradaic constant, z_i is the valence charge of the considered ionic species, D_i is the diffusion coefficient, c_i is the concentration of ionic species, $\mu_{m,i}$ is the effective mobility of the ionic species, ϕ_i is the electrolyte potential, and the i sub-index refers to contribution from a specific ionic species. We assumed that there is no electrolyte flow and thus the current transport is only due to ionic charge's electric field driven transport (migration) and diffusion. We considered sulphuric acid (H_2SO_4) as the electrolyte at various pH , with only H^+ and HSO_4^- ions in the electrolyte. Transport of ionic species through the ion exchange membrane were assumed to follow the same equation but for Nafion's corresponding diffusion coefficients. The ionic species concentration in the electrolyte follows the electroneutrality condition $\sum_{i=1}^n z_i c_i = 0$. In our calculations, the electrodes were considered to be perfectly conducting, the gas species were considered to be immediately extracted upon generation at the electrodes and crossover effects were ignored. We always ensured that the top and bottom computational domain boundaries were sufficiently far away such that there was zero current at these boundaries. The EC current was calculated by integrating the reaction current density at the anode or cathode active surface and normalizing it to the total surface, which includes the membrane area where no water splitting reaction occurs. All relevant physical constants were taken from the contribution of Haussener et al. [14].

The thin-film silicon solar cells were deposited on glass carriers in a medium-sized ($40 \times 50\ \text{cm}^2$) single-chamber plus load lock R&D reactor by plasma enhanced chemical vapor deposition (PECVD). All solar cells were made in a p-i-n configuration, meaning that the respective layers (p, i and n) were deposited in that order. The single and tandem solar cells were used in a superstrate configuration while the triple junction solar cells were used in the substrate configuration. The absorber layers consisting of hydrogenated amorphous silicon (a-Si:H) have a thickness of $500\ \text{nm}$ for the single junction cells while for the tandem and triple junction cells the a-Si:H thickness is $130\ \text{nm}$ and $500\ \text{nm}$ for the top and second layers, respectively. The microcrystalline silicon cells ($\mu\text{-Si:H}$) have an absorber layer thickness in excess of $1\ \mu\text{m}$. All layers were deposited from a gas mixture including SiH_4 and H_2 . For the p-type layers, trimethylborane ($\text{TMB} = \text{B}(\text{CH}_3)_3$) was used as the dopant gas, whereas for the n-type layers phosphine (PH_3) was used. The $2\ \mu\text{m}$ thick transparent conductive contacts consist of boron doped zinc oxide (ZnO:B), and were fabricated by low pressure chemical vapor deposition (LPCVD) in a home-made system from a gas mixture of diethyl-zinc ($\text{Zn}(\text{C}_2\text{H}_5)_2$), H_2O and diborane (B_2H_6).

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