



A monolithic all-silicon multi-junction solar device for direct water splitting

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ABSTRACT

We present a silicon-based, monolithic multi-junction solar device that is suitable for the sustainable and reliable production of hydrogen. It is based on an interdigitated back-contact (IBC) solar cell which is modified, so that the p- and n-regions are connected in a combination of series and parallel connections, which triples the photovoltage compared to a single-junction cell. Thus, it provides a potential larger than the water redox potential of 1.23 V plus over-potentials at the electrodes. We fabricated a working demonstrator with an open-circuit voltage of 1.81 V and a short-circuit current density of 12.2 mA/cm². The processing can be integrated in an existing IBC cell line with one additional fabrication step and a modified contact layout. Coupled to a 1 M H₂SO₄ electrolysis system, with Pt and RuO₂ electrodes, our device shows a solar-to-hydrogen conversion efficiency of 8%. In contrast to existing solutions, which are based on external series connections, it does not suffer from ohmic losses and holds the potential to reach 16.6%.

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1. Introduction

The need for sustainable and reliable production of hydrogen as a regenerative fuel has been a major topic in scientific research for years. In 1995 Bard and Fox defined the “holy grail” of solar water splitting, using the terrestrial AM1.5 spectrum, to split water in H₂ and O₂ with an efficiency of at least 10% and a lifetime of at least 10 years [1]. 20 years later, the required combination of stability, scalability, cost- and conversion-efficiency is still not satisfactory solved. The coupling of photovoltaic and electrolysis (PV-E) systems, however, has the potential to work efficiently and to solve the storage need for excess-peaks in the PV power production [2].

To split water into hydrogen and oxygen it is necessary to provide the redox potential of water of 1.23 V and to additionally compensate over-potentials at the electrodes, which depends mainly on the electrode material, geometry, the choice of electrolyte, and catalysts, as well as the desired current densities [3]. In general, an overall operating voltage of 1.5–2 V is required [4],

leaving two main possible approaches for solar based systems.

The first approach utilises a single material with a high band gap E_G , as the diode saturation decreases exponentially with a wider E_G , and therefore increases the open circuit voltage V_{OC} of a solar cell. A high band gap, however, reduces the light absorption, yielding a small short-circuit current density J_{SC} , and therefore a low hydrogen production.

The second approach employs multiple solar devices connected in series to form a tandem device providing high quantum efficiencies as well as high V_{OC} . An AlGaAs/Si multilayer tandem cell has been demonstrated by Licht et al. that exhibits a high solar-to-hydrogen conversion efficiency (STH) of 18.3% but relies on a cost-intensive fabrication and utilises toxic and rare materials [5].

For a large-scale application a solution based on the well-established, cheap and abundant Si technology is preferable. Urbain et al. have demonstrated low-cost amorphous silicon (a-Si) thin-film tandem cells reaching an STH of 6.8% [6]. The electrical and optical properties of a-Si, however, are known to be strongly dependent on the thermal history of the amorphous solid. Zellama et al. have shown an irreversible change of the optical gap and the defect density, starting at about 300 °C induced by a change in the microstructure and hydrogen content [7]. This thermal sensitivity

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restricts the application of promising new catalysts like manganese oxides and molybdenum sulfides that require processing temperatures up to 500 °C [8,9]. Another recent low-cost application relies on the rather new type of perovskite solar cells. Tandem structures showed a promising STH of 12.3% but are not long-time stable [10].

A series connection of crystalline Si (c-Si) cells offers a possibility to overcome the thermal limitations at moderate costs. Gibson and Kelly used state-of-the-art PV modules consisting of multiple series and parallel connected solar cells coupled to an external proton exchange membrane electrolyser stack. With this setup, an STH of 12.4% is reached [11]. Due to the high-power PV modules, a pumped cooling system for the electrolyte was required, which is not part of the efficiency calculations. The induced circulation of the electrolyte removes evolving gas bubbles from the electrodes, making the system more efficient compared to static electrolyte realisations. The major issue of PV modules, however, is the reduction of the fill factor (FF) due to higher series resistances R_S caused by additional soldering points and interconnections [12]. This ohmic loss can be reduced by connecting halved cells in series and, therefore, halving the current density and decreasing the ohmic losses [13]. A mechanical halving has to address side-edge passivation and may lead to wafer breakage. Cutting the solar wafers prior to cell processing necessitates a new process line to handle the smaller wafers. Therefore, a monolithic solution is desirable, employing a highly efficient Si solar cell, manufactured by a well-established process, that is modified to a minimum to ensure the necessary series connection.

We present an approach, in which a monolithic Si multi-junction will directly seal an electrolyte container and can be either run with a mechanical gas separation, or equipped with an additional proton exchange membrane. A working triple-junction is fabricated in a top-down process, modifying an existing interdigitated back contact (IBC) cell. The impact of these modifications on the PV characteristics are discussed and analysed using the two-diode model. To estimate an upper limit for a device based on the given single-junction IBC cells, we eliminate the lumped recombination at the front- and back-side in the two-diode model, adapting parameters obtained from single-junction measurements. This ideal triple-junction represents the theoretical result of a tailored fabrication process based on the given IBC cells. Coupled to an electrolysis system our fabricated triple-junction shows a STH efficiency of 8% and has the potential to reach 16.6%.

2. Monolithic multi-junction PV-E concept

The multi-junction PV-E device, as schematically shown in Fig. A.1, is based on the IBC solar cell concept, that offers a strict spatial separation between the light in-coupling front-side and the charge carrier collection on the back-side. A single IBC solar cell consists of several p- and n-regions (called fingers) that are connected in parallel to reach a V_{OC} of roughly 700 mV [14]. Therefore, three equal cell areas are connected in series, forming a triple-junction IBC cell device, with a V_{OC} suited to the electrolysis operating voltage of 1.5–2 V. The series connections are formed monolithic on top of the p- and n-junctions at the back-side of the solar cell wafer (Fig. A.2). Simulations using SENTAURUS TCAD predict that the series connection of the cells works only with a deep trench isolation (DTI) on the front-side, isolating the single cell areas (Fig. A.3). A depth of at least about 171 μm is vital for a 180 μm thick substrate (i.e. 95%) to obtain a high V_{OC} , while J_{SC} is almost not affected. It is, therefore, a crucial part of this concept to find a trade-off between mechanical stability and depth of the trenches. The back-side of the triple-junction cell will be covered with a thick, inert silicon dioxide or nitride, thus, passivating the PV system against corrosion in the harsh electrolysis condition. The

electrolysis contacts will be formed on top of the passivation layer and matched to the current density at the operating point of the PV-E system enhancing the overall efficiency. Besides the reduction of ohmic losses, this concept does not suffer from light absorption in the electrolyte and allows a direct heat transfer from the illuminated PV to the electrolysis system, making both more efficient.

3. Experimental

3.1. Fabrication of monolithic triple-junction IBC cells with DTI

IBC cells provided by IMEC [14], with a size of $2.5 \times 2.5 \text{ cm}^2$ and a thickness of about 150 μm , are modified in order to demonstrate the multi-junction cell concept as described above. The Al (1% Si) busbars are segmented using a photolithography process with a subsequent PAN and reactive ion etching (RIE) step. The monolithic series connection is formed by applying a photolithography step followed by an electron-beam evaporation of 200 nm Al and a lift-off process. The DTI on the front side is defined by a resist and photolithography step and a deep RIE process (Bosch process). Further details can be found in the supplementary materials.

3.2. Measurement systems

The electrical characterisation was carried out with a HP 4156A Precision Semiconductor Parameter Analyzer (Keysight), a self-made mounting frame and an Oriel3A class AAA sun simulator (Newport) with an AM 1.5 air mass filter, together with a calibrated reference cell. The measured current of the single-junction IBC cells at 1 sun exceeds the power limit of the HP 4156A. Therefore, the single-junction measurements are taken at 0.25 suns, whereas the triple-junction cells are measured at 1 sun. The temperature is measured within the reference cell. The time of illumination is kept constant and as short as possible, utilizing a shutter in the sun simulator. Measurements are taken prior and after processing of the IBC cells. The measured current density voltage (J – V) characteristics are fitted by using a MATLAB based two-diode modelling tool [15,16]. To further validate our measurement, finite element simulations of the IBC cells are carried out with SENTAURUS TCAD. For the electrolysis measurements, the cells are mounted in the contacting frame and are externally wired to the electrolysis system. 1 M H_2SO_4 serves as electrolyte, solid Pt as the cathode, for the production of hydrogen, and 100 nm of ruthenium dioxide (RuO_2) as the anode for the oxygen evolution. The RuO_2 is reactively sputtered on top of 100 nm electron-beam evaporated Ti on a 2.4 cm^2 glass substrates.

The whole setup is shown schematically in Fig. A.4. The separation between PV and electrolysis is needed to introduce the electrical measurement in between PV and electrolysis system. Future work will address a measurement of the evolved gasses for a non-wired system as presented in Fig. A.1. The operating point of the PV-E system is determined by connecting this two-electrode setup to the HP 4156A Analyzer, which is set to measure the voltage between the electrodes and the current flowing from the cathode to the negative pole of the IBC cell. The current density as well as the electrode potential is measured time-resolved under illuminated and dark conditions. More information on the measurement system are given in the supplementary materials.

4. Results and discussion

The first part of the following section gives a detailed analysis of the PV system. The impacts of the structural modifications on the single-junction cell characteristics are evaluated before the results of the monolithic coupled triple-junction are presented. Based on

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