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Light-induced degradation of adapted quadruple junction thin film silicon solar cells for photoelectrochemical water splitting

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ABSTRACT

The fabrication process of high performance quadruple junction thin film silicon solar cells is described and the application of the solar cells in an integrated photoelectrochemical water splitting device is demonstrated. It is shown that the performance of solar cells can be adjusted by varying the process parameters and the thickness of the absorber layers of the individual sub cells and by integrating microcrystalline silicon oxide as intermediate reflecting layers. Thereby current matching of the sub cells was improved and a high open-circuit voltage of 2.8 V was achieved. Furthermore, the solar cell stability against light-induced degradation was investigated. Efficiencies of 13.2% (initial) and 12.6% (after 1000 h of light-soaking) were achieved. Bias-free water splitting with a solar-to-hydrogen efficiency of 7.8% was demonstrated in an integrated photovoltaic–electrochemical device using the developed quadruple junction photocathode. Finally, it is shown that in the case of quadruple junction solar cells the light-induced degradation has a lower effect on the photovoltaic–electrochemical efficiency as on the photovoltaic efficiency.

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1. Introduction

Multijunction thin-film silicon solar cells offer improved utilization of solar spectrum and reduced thermalization losses in comparison to single junction devices [1]. Tandem and triple junction configurations of solar cells based on hydrogenated amorphous silicon (a-Si:H) and microcrystalline silicon ($\mu\text{c-Si:H}$) have been extensively studied, showing high initial efficiencies above 13.5% [2] and 16.3%, respectively [3]. Theoretical studies demonstrate that an increase in the number of sub cells in a multijunction device results in an improved efficiency [4], and recently, the development of quadruple junction thin film solar cells has been a subject of research in several groups [5–7].

Here, we present a development of quadruple junction thin film silicon solar cells for the application in an integrated water-splitting device for hydrogen production. Photoelectrolysis of water by semiconductor based devices represents a prominent route to store electrical energy in the form of a clean chemical fuel, namely hydrogen and recently raised increasing interest among research groups worldwide [8–10]. Solar assisted water splitting

requires photovoltages above 1.8 V. Therefore, monolithic multi-junction thin film silicon solar cells are very promising candidates to be applied as a power source for water splitting [11–13]. However, the exact voltage needed strongly depends on the catalyst materials used to enhance the water splitting reactions [14]. Despite considerable efforts in the development of cost-efficient catalysts, precious metal catalysts still require lower voltages than cheaper and earth-abundant catalyst materials [15]. Thus, in order to have a high flexibility in choosing abundant catalyst materials high open circuit voltages are desirable along with good photovoltaic efficiencies.

We focus on both high initial and stabilized efficiencies of the quadruple solar cell. Our approach here is to avoid utilization of alloyed absorber materials, such as a-SiO_x:H or a-SiGe:H [5,6], due to known stability issues under prolonged illumination. Thus, we apply intrinsic a-Si:H and $\mu\text{c-Si:H}$ absorber layers. For a-Si:H a variation in the bandgap is achieved by changes in the deposition temperature and/or gas flow ratios as previously presented for the case of tandem devices [16,17]. Additionally, we utilize doped $\mu\text{c-SiO}_x\text{:H}$ layers [18] to improve the current transfer between sub cells and the current matching in the quadruple junction device. The optimized quadruple junction solar cell shows high efficiencies of 13.2% (initial) and 12.6% (stabilized). The application of

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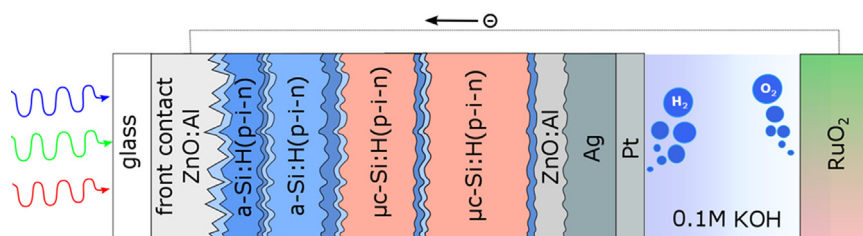


Fig. 1. Schematic illustration of the PV-EC device configuration investigated in this study, with its components: the multijunction photocathode (a-Si:H/a-Si:H/ μ c-Si:H/ μ c-Si:H), Pt as HER catalyst layer at the photocathode/electrolyte interface, the electrolyte (0.1 M KOH), and the RuO₂ anode as OER catalyst. Hydrogen evolves at the rear side of the photocathode and oxygen evolves at the RuO₂ anode.

Table 1

Overview of the used light wavelengths for the saturation of the individual sub cells in quadruple junction solar cells for the QE evaluation.

Cell type	Measured sub cell			
	Top cell (nm)	Middle cell 1 (nm)	Middle cell 2 (nm)	Bottom cell (nm)
a-Si:H/a-Si:H/ μ c-Si:H/ μ c-Si:H	590	780 and 400	525 and 830	625

the quadruple solar cell in an integrated photovoltaic–electrochemical (PV-EC) device is demonstrated with a bias-free solar-to-hydrogen (STH) efficiency of 7.8%. Apart from the STH efficiency, the performance of photoelectrochemical devices is also evaluated with respect to their stability. In thin film silicon based PV-EC devices two major degradation mechanisms are particularly present: the electrochemical and the light-induced photovoltaic degradation. Herein, we address the impact of the light-induced photovoltaic degradation on the PV-EC performance also in comparison to the solar cell performance degradation.

2. Experimental

2.1. Preparation of thin film silicon layers and solar cells

All thin film silicon layers were deposited by a plasma enhanced chemical vapor deposition technique in a multi chamber system. The solar cells were deposited on 100 cm² textured aluminum-doped zinc oxide (ZnO:Al) coated glass substrates (front contact) [19]. For the intrinsic absorber layers a mixture of silane (SiH₄) and hydrogen (H₂) gases was used. For the p- and n-type layers, trimethylborane (TMB), methane (CH₄), carbon dioxide (CO₂) and phosphine (PH₃) gases were added to the silane-hydrogen mixture. An excitation frequency of 94.7 MHz was applied for the deposition of the μ c-Si:H p-type and the intrinsic layers. For all a-Si:H layers an excitation frequency of 13.56 MHz was applied. The solar cells were fabricated in p-i-n superstrate configuration with a sputtered aluminum doped zinc oxide/silver reflecting rear contact [19] defining the area (1 cm²) of the individual cells. For all intrinsic μ c-Si:H absorber layers a silane concentration (SC) of 5.0%, defined as the ratio between the SiH₄ flow and the total gas flow, and a substrate deposition temperature (T_S) of 180 °C was chosen. The μ c-Si:H absorber layers were deposited at an average rate of 0.37 ± 0.03 nm/s and had a crystalline volume fraction, evaluated from Raman measurements [20] of around 50%. Additional details on the preparation conditions of μ c-Si:H absorber layers can be found in Ref. [17]. The intrinsic a-Si:H top and middle cell absorber layers were deposited at 130 °C with a SC of 4% and at 180 °C with a SC of 10%, respectively. The optical band gaps of the corresponding a-Si:H absorber layers, measured by Photothermal Deflection Spectroscopy [16] are $1.95 \text{ eV} \pm 25 \text{ meV}$ and $1.91 \text{ eV} \pm 25 \text{ meV}$, respectively.

We used μ c-SiO_x:H n-layers and integrated an intermediate reflecting (IR) layer [18] between the middle a-Si:H and the first μ c-Si:H sub cell, as schematically depicted in Fig. 1. In order to match the sub cells in terms of the highest possible photocurrent, the thicknesses of each cell were systematically adjusted.

2.2. Current–voltage and quantum efficiency measurement of quadruple junction solar cells

The solar cells were characterized in the initial and stabilized states (after 1000 h light-soaking) by current–voltage (*J*-*V*) measurements at standard test conditions (100 mW/cm², 25 °C) using a double source (Class A) AM 1.5 sun simulator with an anti-reflection foil on top of the front glass [21]. Light-induced degradation (LID) was performed under open-circuit conditions at 55 °C under simulated sun spectrum (1000 W/m²) during 1000 h.

Spectral response measurements [external quantum efficiency (QE)] of the quadruple junction solar cells were conducted before and after light-soaking. A monochromator was used to select individual wavelengths in the range between 300 nm and 1100 nm. The individual sub cell QEs of the quadruple junction solar cells were separately determined using LEDs and a spectrally filtered bias light from a tungsten lamp. The corresponding wavelengths used for the saturation of the sub cells were determined empirically and are presented in Table 1.

To measure the photocurrent density of the top cell, light with 590 nm wavelength was used in order to saturate both middle cells and the bottom cell. In this case, the top cell, whose photocurrent was intended to be measured, limits the current of the whole device. For the measurement of the bottom cell, light of 625 nm wavelength was used to saturate the top and both middle cells. In the case of the first middle cell QE measurement, the top cell is saturated with a 400 nm light and both the second middle cell and the bottom cell are flooded with 780 nm light. To measure the photocurrent of the second middle cell, a wavelength of 525 nm was used to saturate the top and the first middle cell and light of 830 nm wavelength ensures that the bottom cell is not the current limiting cell.

2.3. Photoelectrochemical measurements

The photoelectrochemical performance of the photocathode was evaluated at room temperature in an aqueous 0.1 M potassium hydroxide (KOH) solution by means of linear sweep

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