



TECHNOLOGY DEMONSTRATION OF LARGE-SCALE PHOTO-ELECTROCHEMICAL SYSTEM FOR SOLAR HYDROGEN PRODUCTION

Project Deliverable Report – D3.5

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EXECUTIVE SUMMARY

The purpose of the report is to present the recent test measurements for the 3rd generation of the thermally integrated CIGS-alkaline electrolyser with an active device area of $\sim 100 \text{ cm}^2$. **The main objective** of the deliverable was changed from performing a 1000 h test with a prototype of an integrated CIGS-electrolyser and outdoor test of an upscaled device to reporting performance of the final prototype device and outdoor test results. The reason for this change was the departure from the project, of the industrial partner Solibro Research that would have provided large area CIGS modules.

The thermally integrated photovoltaic (PV)-electrolyser with a 78 cm^2 area of 4-cell CIGS module showing 16.1 % PV efficiency under 100 mW cm^{-2} illumination and an electrolyser with 100 cm^2 NiMoV (cathode)-NiO(anode) thin-film catalysts achieved a 9.1 % maximum and 8.5 % averaged solar-to-hydrogen (STH) efficiency for 100 hours. The average hydrogen production rate of 3.3 ml min^{-1} and area-specific hydrogen production rate of $2.75 \text{ g h}^{-1}\text{m}^{-2}$ were reported in **deliverable 3.3**.

In **deliverable 3.4**, we reported 10% and 9.7 % maximum and average STH efficiency for a 7-day measurement, corresponding to 5.4 ml min^{-1} average hydrogen production rate, and $2.87 \text{ g h}^{-1}\text{m}^{-2}$ area-specific hydrogen production rate for a 2x3-cell CIGS module having 14.3 % efficiency integrated to a FeNi layered double hydroxide (LDH) catalysts-based electrolyser where the catalysts and the PV module had 100 cm^2 areas.

In this report, **deliverable 3.5**, 100 cm^2 FeNi DLH catalysts prepared by a hydrothermal method on Ni foam were used as cathode and anode material of the alkaline electrolyser. The CIGS module consisted of 2x3-cells with an active total area of 82.32 cm^2 and had a module efficiency of 17.27 % at $25 \text{ }^\circ\text{C}$ under 100 mW cm^{-2} illumination. The device design consisted of the following stack: PV module / Ni plate / cathodic catalyst on Ni foam / gasket/ membrane /gasket / anodic catalyst on Ni foam / Ni plate.

The maximum STH efficiency determined from the gas volume measurement was recorded as 13.4 % under 1000 W/m^2 irradiance. The average STH efficiency was 11.3 % with the corresponding average hydrogen production rate of 5.74 ml min^{-1} and area-specific hydrogen production rate of $3.74 \text{ g h}^{-1}\text{m}^{-2}$.

The thermally integrated device has been placed at the installation site in Julich in November 2020, Germany, for the outdoor test. Although, the weather has not been good and the intensity of the sun has been relatively low, it was reported that the system has been running well. The preliminary data showed that STH efficiency was in the range of 10 %. The outdoor test for the system continues.

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ABBREVIATIONS and ACRONYMS

CA	Consortium Agreement
CIGS	CuInGaSe
CNR	Consiglio Nazionale delle Ricerche
DLH	Double layer hydroxide
DoA	Description of Action
EC	Electrolyser
EGP	Enel Green Power S.p.A
ETH	Electricity-to-hydrogen
FZJ	Forschungszentrum Jülich GmbH
GA	Grant Agreement
HER	Hydrogen evolution reaction
HZB	Helmholtz Zentrum Berlin
I-V	Current-voltage
I_{mp}	Current at maximum power point
I_{sc}	Short circuit current
LSV	Linear sweep voltammetry
MS	Milestone
OER	Oxygen evolution reactions
PV	Photovoltaic
SRAB	Solibro Research AB
STH	Solar-to-hydrogen
T	Temperature
TRL	Technology Readiness Level
I_{int}	Current at intersection
UU	Universität Uppsala
V_{int}	Potential at intersection
V_{mp}	Potential at maximum power point
V_{oc}	Open circuit potential
WP	Work Package
XRD	X-ray diffraction
η	PV module efficiency

1 INTRODUCTION

The purpose of this report is to present the recent test measurements for the 3rd generation thermally integrated CIGS-alkaline electrolyser with a 100 cm² area.

In deliverable 3.3, we had obtained 9.1 % maximum and 8.5 % averaged STH efficiency for a thermally integrated PV-electrolyser having a 4-cell CIGS module with a 78 cm² active area and 16.1 % efficiency under 100 mW cm⁻² illumination and an electrolyser with 100 cm² NiMoV (cathode)-NiO(anode) thin-film catalysts for 100 hours. The average hydrogen production rate was 3.3 ml min⁻¹.

We rebuilt the device with a CIGS PV module to improve the hydrogen production rate, which gave a better matching operation current and FeNi layered double hydroxide (LDH) catalysts in the electrolyser, which had less overpotential than the thin film catalyst (D3.4). Here, we used a 100 cm² CIGS module with two parallel connected units of 3 interconnected cells in a resulting 14.3 % area PV efficiency. The 10% and 9.7 % maximum and average STH efficiency were obtained for a 7-day measurement, respectively and the average hydrogen production rate was improved to 5.4 ml min⁻¹.

The main objective of deliverable D3.4 was to perform a 1000 h test with the integrated CIGS-alkaline electrolysis systems. However, the full 1000 h could not be reached due to the leakage of the electrolyser, which destroyed the PV module as well. The task of deliverable 3.5 was to report the outdoor test results of an upscaled CIGS-electrolyser. However, as one of the key partners in this deliverable, SRAB, left the project due to insolvency, the target of deliverable 3.5 was reformulated. The new aim was to fix the problems in the existing setup and to perform the outdoor tests on the prototype. The work for deliverable 3.5 was led by WP3 (UU) and done in collaboration with SRAB and FZJ. SRAB provided new modules before they left the project. UU produced and characterized electrocatalysts, fixed the leakage issue, and rebuilt the PV-electrolyser device. In the emerging COVID-19 situation, it was not possible to send a person from UU to FZJ to set up the device in Jülich. The device was posted to Jülich and FZJ installed the device in the outdoor test bed in November 2020 and did the performance measurements.

2 DELIVERABLE OBJECTIVE AND RELATED TASKS

The deliverable objective is to report on efficiency and hydrogen production, and the feasibility of the CIGS-alkaline electrolyser prototype device (100 cm²) which was sent to FZJ for the outdoor test.

2.1 Description of the task

No.	Task description	Start date	End date
T 3.3	10-30 cm ² Alkaline water splitting modules	Mar 2018	Dec 2020

T3.3 focused on transferring knowledge and up-scaling of the CIGS modules in two steps, up to 10 cm × 10 cm, and then realizing the concepts in larger modules totaling 10 m². Here, an investigation of the PV-electrolyser was done in co-operation with SRAB and FZJ. The resulting data is used in WP6 for cost-analysis and life-cycle analysis for the CIGS and catalysts for larger devices totaling 10 m².

FROM THE GA – DoA:

T3.3 will focus on transferring knowledge and up-scaling of the CIGS modules in two steps; up to 10 cm × 10 cm and then realizing the concepts in larger modules totaling 10 m². Patterning techniques for the PV part will primarily be based on laser methods, but mechanical scribing and photolithography-based methods will be second alternatives. Here, catalyst modules in co-operation with SRAB and

cassette design with CNR, and the joint analysis of sealing approaches lead by HZB will be investigated. In WP3.3, the lead partner is SRAB with support and interaction from UU, HZB, CNR, and also interconnected and analyzed in context to the Si approach from FZJ, HZB, and EGP. The last WP will also include cost-analysis and life-cycle analysis for the CIGS and catalysts in prolonged use and support WP7.

3 CONTRIBUTION TO PROJECT OBJECTIVES

This deliverable contributes directly and indirectly to the achievement of main and specific objectives indicated in section 1.1 of the Description of the Action (GA -735218):

<i>Project Objectives</i>	<i>Contribution of this deliverable</i>	
	<i>Yes</i>	<i>No</i>
Main objective		
To demonstrate an operational PV-EC system measuring at least 10 m ² with a solar to hydrogen (STH) efficiency of at least 6 % supporting a hydrogen production of at least 16 g/h at a levelised cost of 5€/kg	✓	
Specific objectives		
To study and develop devices for integrated PV-EC concepts and scale viable concepts to prototype size > 100 cm ²	✓	
To use socio-techno-economic analysis to predict and select concepts with levelised cost of hydrogen production below € 5/kg	✓	
To scale the prototypes of the less mature but promising technologies to a demonstrator with active area > 10 m ²	✓	
To achieve a hydrogen production of 16 gH ₂ /h from the demonstrator resulting in a STH efficiency of at least 6 %	✓	
To ensure that the initial demonstrator STH efficiency does not reduce by more than 10 % after six months of continuous operation	✓	

4 DESCRIPTION OF COMPLETED ACTIONS

4.1 EC characterization

In this report, 100 cm² FeNi DLH catalysts, hydrothermally grown on Ni foam, were used as cathode and anode material of the alkaline electrolyser for the solar hydrogen production, in contrast to the previous generations of the device where the catalyst layers were deposited with sputtering.

The FeNi DLH catalysts were prepared by a hydrothermal method that is scalable and would allow deposition of larger areas in the future. The catalyst surface morphology is shown in Fig. 1a. X-ray diffraction (XRD) (Fig. 1b) and Raman spectroscopy (Fig. 1c) characteristics of the pristine catalyst were consistent with the previous work¹. The catalysts were activated in 1.0 M KOH by applying a current corresponding to 10 mA cm⁻² for 100 h at 25°C. After the activation, the overpotential of hydrogen evaluation reaction (HER) (Fig. 1d) and oxygen evaluation reaction (OER) (Fig. 1e) was decreased from 262 to 189 mV and 212 to 201 mV, respectively.

¹ Qiu, Z., Tai, C.-W., Niklasson, G. A., and Edvinsson, T. (2019). Direct observation of active catalyst surface phases and the effect of dynamic self-optimization in NiFe-layered double hydroxides for alkaline water splitting. *Energy & Environmental Science* 12, 572-581.

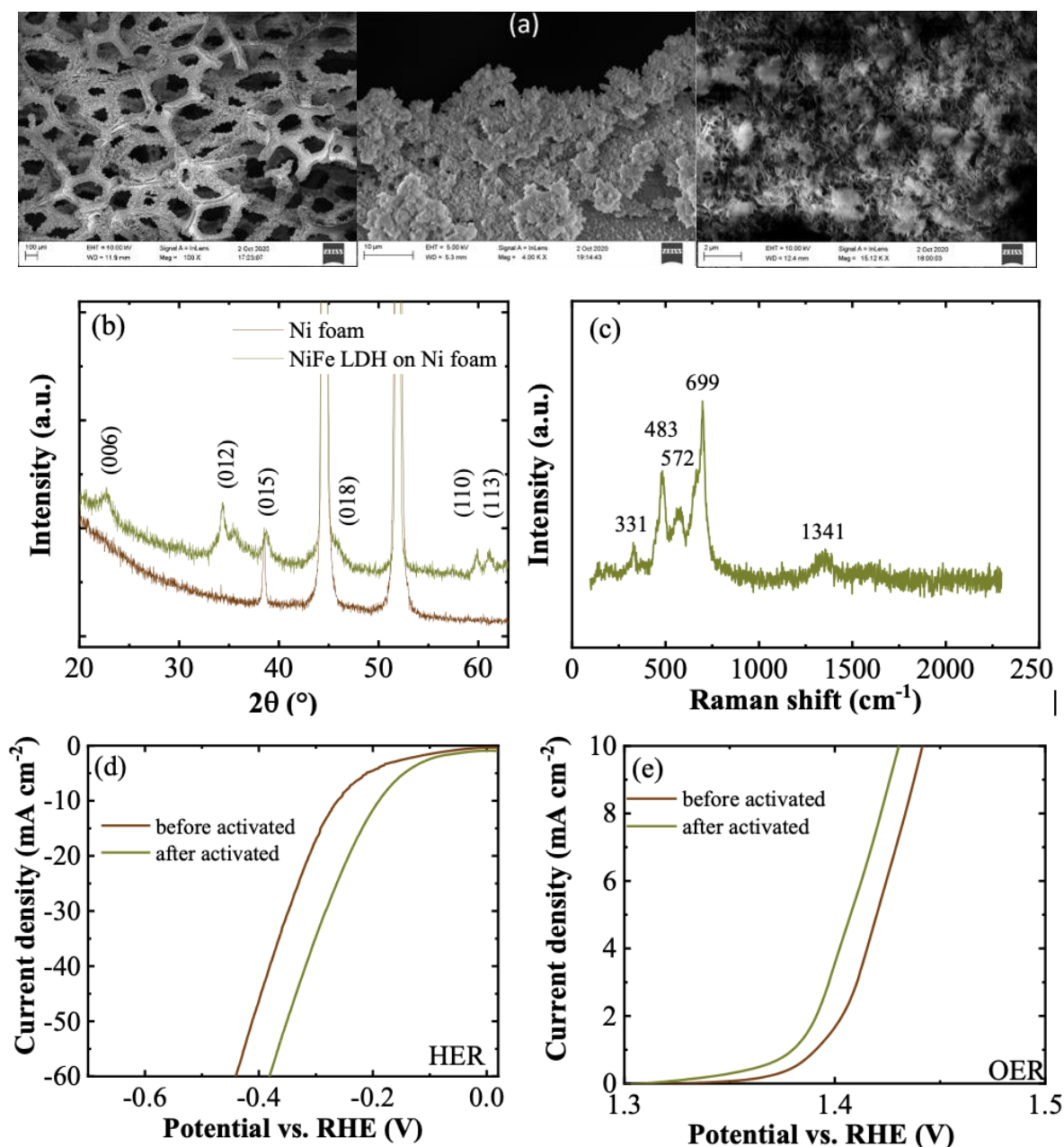


Figure 1. SEM images (a), XRD pattern (b), Raman spectrum of as-prepared FeNi LDH catalyst formed on Ni foam. Linear sweep voltammograms for HER (d) and (OER) of the FeNi LDH catalyst in 1.0 M KOH at 25°C with a scan rate of 5 mV s⁻¹.

4.2 PV characterization

In this deliverable, a CIGS module was developed using 2 parallel cell strings, each consisting of 3 monolithically series interconnected cells giving a total active area of 82.32 cm². The module PV efficiency decreased from 17.27 to 16.06 % (Fig. 2a) when the temperature increased from 25 to 55 °C, and the fill factor (*FF*) was below 68 % (Fig. 2b). Under 100 mW cm⁻² illumination, the module's open-circuit voltage (*V*_{oc}) decreased from 2.27 to 2.12 V, with temperature increasing from 25 to 55 °C (Fig. 2c). The short circuit current (*I*_{sc}) at 902 mA was largely unaffected by varying operating temperature from 25 to 55 °C (Fig. 2d).

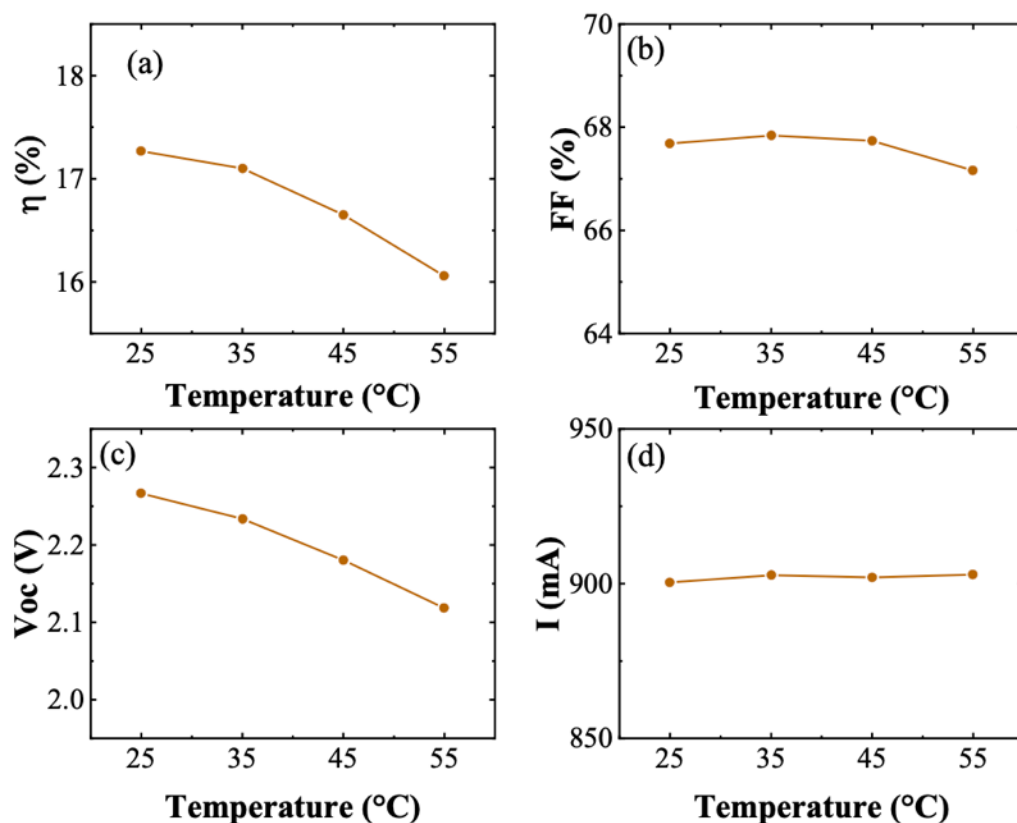


Figure 2. Effect of module temperature on PV module efficiency (a), fill factor (b), open circuit potential (c), and short circuit current (d) for the CIGS module.

4.3 Device design

The device design and a photograph of the integrated CIGS-electrolyser are shown in Figures 3a and 3b, respectively. The electrolyser part consists of Ni plate / cathodic catalyst on Ni foam / gasket/ membrane / gasket/ anodic catalyst on Ni foam / Ni plate stack. Figure 3c shows a photograph of the setup during current-voltage (I - V) measurements of the electrolyser and Figure 3d and 3e photographs from measurements of CIGS and hydrogen volume in Jülich. All measurements were done with active flow via pumping. The pumps forced electrolyte circulation between the electrolyser and reservoirs through Teflon tubes with a flow rate of 50 ml min^{-1} . The volume of the photogenerated gas was measured using inverted graduated cylinders with 2 ml accuracy.

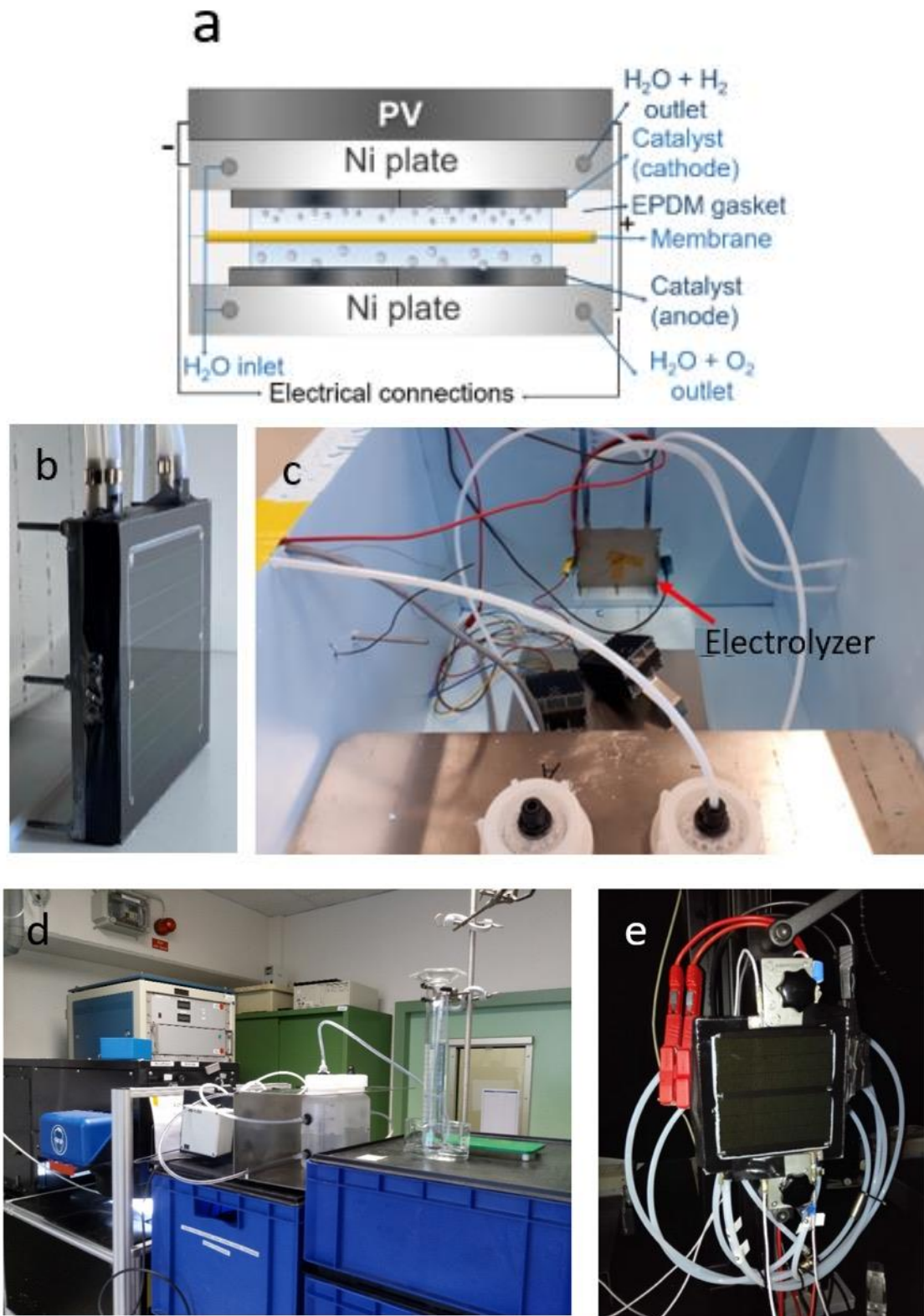


Figure 3. Schematic picture (a) and a photograph (b) of the integrated CIGS-electrolyser , a photograph of the setup at UU (c), and pictures from the indoor measurements in Jülich (d and e).

4.4 Current-voltage data of the CIGS and electrolyser

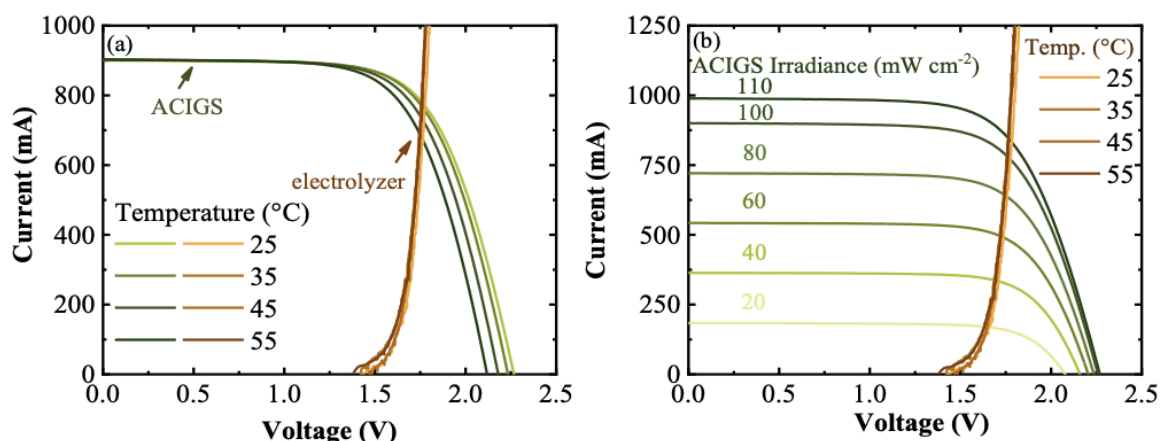


Figure 4. The experimental I-V data of alkaline FeNi LDH (cathode)-FeNi LDH (anode) electrolyser with a catalyst area of 100 cm² and a CIGS module consisting of 2×3 cells (a) at an irradiance of 100 mW cm⁻² and temperatures between 25°C and 55°C (b) with varying irradiance upon the CIGS module (temperature fixed to 25°C) and the varying temperatures for the electrolyser.

The catalyst's experimental data were obtained at UU at different temperatures between 25 and 55 °C (Fig. 4a and 4b). I-V measurements of the CIGS module were done in FZJ at the same temperature range (Fig. 4a) and at different irradiance (Fig. 4b). STH efficiency of the CIGS module-electrolyser was calculated from the intersection current of the module and electrolyser. The STH efficiency decreased from 11.6 to 10.3 % with increasing temperature (Table 1) due to the drastic changes in PV current after the maximum power point. This calculation was done assuming that the PV and electrolyser are at the same temperature.

On the other hand, temperature changes in the electrolyser did not significantly affect the catalytic current. The results thus indicate that one would not expect a noteworthy difference in STH efficiency. As an example, if the PV temperature is 55 °C and electrolyser temperature is any temperature between 25 and 55 °C, the STH efficiency only changes from 10.1 to 10.3 %.

Table 1. Operation potential (V_{int}), current (I_{int}) and STH efficiency determined from intersection of I-V curves of 6 cells CIGS and alkaline FeNiOH (cathode)-FeNiOH (anode) electrolyser for the same temperatures for PV and electrolyser with an irradiance of 100 mW cm⁻².

T (°C)	V_{int} (V)	I_{int} (mA)	STH (%)
25	1.77	772	11.6
35	1.76	766	11.5
45	1.75	739	11.1
55	1.75	690	10.3

The STH efficiency increased from 11.4 to 12.7% with decreasing irradiance (Table 2); arising from the lower required overpotential at lower catalytic current and a more optimally placed catalyst load curve with respect to the PV IV-curve. However, since the hydrogen production is proportional to the current, one expects a lower amount of hydrogen at lower irradiance since it decreased from 834 to 170 mA with decreasing irradiance from 110 to 20 mW cm⁻², respectively.

Table 2. Operation potential (V_{int}), current (I_{int}) and STH efficiency from intersection of I-V curves of 2×3-cell CIGS subjected to different irradiances and alkaline FeNi LDH (cathode)-FeNi LDH (anode) electrolyser .

Irradiance (mW cm^{-2})	V_{int} (V)	I_{int} (mA)	STH (%)
110	1.78	834	11.4
100	1.77	771	11.6
80	1.76	639	12.0
60	1.74	488	12.2
40	1.71	335	12.6
20	1.66	170	12.7

4.5 Solar hydrogen production measurement in FZJ

The STH efficiency and hydrogen production of the PV-electrolysis device were investigated under laboratory conditions at FZJ before the outdoor test. At the beginning of the measurement, the STH was 8.5% but remained above 10 % for more than 1 hour during an 11-hour continuous monitoring at 100 mW cm^{-2} (Fig. 5). The STH values obtained from hydrogen production agreed with the ones calculated from data (Section 4.4). The maximum STH from the gas volume measurement was 13.4 % (even higher than STH from I - V intersection between the PV and electrolyser), showing that the device design has very low mass transport losses and a Faradaic efficiency close to unity. The average STH efficiency was 11.3 %, with the corresponding average hydrogen production rate of 5.74 ml min^{-1} and an area-specific hydrogen production rate of $3.74 \text{ g h}^{-1}\text{m}^{-2}$.

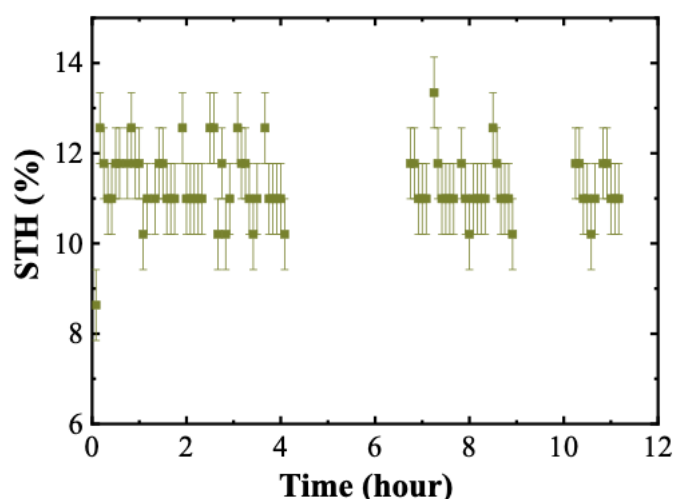


Figure 5. STH efficiency of the integrated PV 2×3 cell CIGS module with a total area of 82.3 cm^2 -alkaline electrolyser (FeNiOH (cathode)-FeNiOH (anode) with 100 cm^2 catalyst area under 100 mW cm^{-2} illumination for 11-hour test measurement. Electrolyte was 1 M KOH .

4.6 PV-electrolysis outdoor measurement in FZJ

In November 2020, the thermally integrated device was installed in the outdoor test site in Jülich, Germany. Although the weather has not been good and the intensity of the sun has been relatively low, the system has been running well. The first outdoor data showed that STH efficiency was in the range 10%. More outdoor data will be collected in the next weeks and reported in deliverable 7.6.

5 DEVIATIONS AND CORRECTIVE ACTIONS

The objective of the deliverable was to perform outdoor measurements in Jülich for an upscaled CIGS-alkaline electrolyser. However, due to the issues described in the introduction in more detail, such as

technical and insolvency of SRAB, the focus of deliverable 3.5 was reformulated to aim for the highest STH efficiency possible with non-precious catalysts and CIGS PV technology, as a corrective action. The new aim was to fix the leakage problems, perform the measurements, and do the outdoor tests on the prototype.

6 DISSEMINATION AND UPTAKE

6.1 Dissemination activities

We recorded our CIGS-electrolyser set-up into a demonstration video with an introduction. The video was published on our institute's (UU) webpage², the PECSYS website, and posted on YouTube via the HZB YouTube channel. Photographs of the device were used for information on the PECSYS website and for other communication tools such as the brochure and the announcement of the project workshop. The results of this device were also presented at the PECSYS project workshop held on 5th November 2020. A manuscript with the latest generation device is under preparation.

As indicated in the Description of the Action (DoA), the audience for this deliverable is:

(PU) – General public	✓
(PP) – Project partners, including the Commission Services	
(CO) – Confidential; only for Consortium Members and the Commission Services	

7 EVALUATION OF THE REPORT FINDINGS

7.1 Comparison with state of the art

A comparison with the results within the project is given in Table 3. In D3.3, 8.5 % (maximum 9.1%) STH efficiency was obtained using a CIGS module with the size of 78 cm²-alkaline electrolysis using NiO/NiMoV thin-film catalysts. In D3.4, 9.7 % (maximum 10 %) STH efficiency was obtained using a CIGS module with the size of 100 cm²-alkaline electrolysis with FeNi LDH nanocatalyst. In the silicon heterojunction (c-Si, SHJ) approach, the catalysts were supported on nickel foam. The electrolyte was 1.0 M KOH, and the data were measured under outdoor conditions 974 W/m² and an ambient temperature of 12.6°C with the PV module at 27.7°C.

Table 3. Comparison of the results in PECSYS for integrated PV-electrolysis.

PV device	OER/HER	Active Area - PV (cm ²)	Area – catalyst (cm ²)	STH (%)	Time (hour)	Area specific hydrogen production rate (g/h/m ²)	Ref.
4 cell CIGS	NiO/NiMoW	78	100	8.5	100	2.75	D3.3
3 cell c-Si (SHJ)	FeNi/Ni	294	50	3.68		1.1	D2.4
6 cell CIGS	FeNi LDH/ FeNi LDH	100.8	100	9.7	100	2.87	D3.4
6 cell CIGS	FeNi LDH/ FeNi LDH	80	100	11	10	3.74	This work
Requirement for MS 3 & MS 4		100	Unspecified	10	1000	1.6	

² <https://materialwetenskap.uu.se/research/solid-state-physics+/news/?tarContentId=908170>

A comparison of the deliverable results with literature data and our previous reported deliverables is shown in Figure 6. Literature data with open magenta-colored hexagon symbols are for PV-electrolysis approaches. The results show that our integrated PV-electrolyser performance is among the largest integrated devices with high efficiency and stability and offering promising scalability.

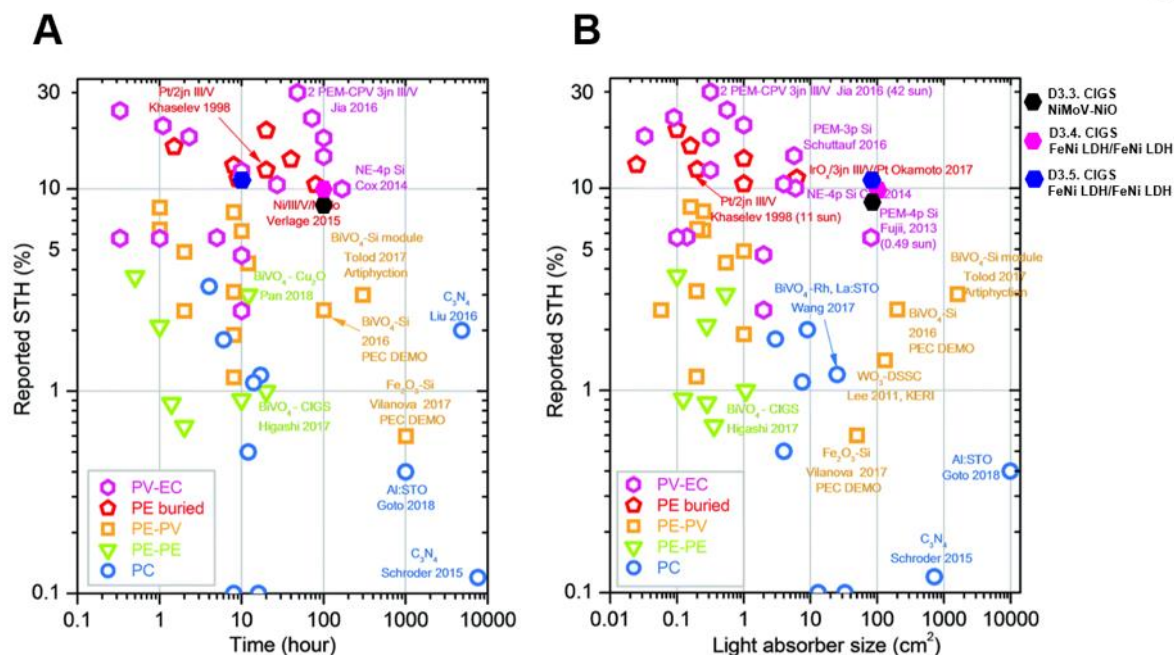


Figure 6. Comparison of the STH efficiency, stability and scalability of the integrated CIGS PV-alkaline electrolyser with the results of deliverable 3.3 and 3.4, and various types of solar hydrogen production systems³.

7.2 Lessons learnt – both positive and negative from the experiences of the work to date

The initial scale-up approach was dependent on an industrial partner (SRAB) that left the project due to insolvency. Therefore, we reformulated the task to instead make a smaller, but still relevant sized, area (~100cm²) prototype for the outdoor tests. This together with the ability to design an integrated PV-EC prototype where the photogenerated gas evolution matched well with the efficiencies from the intersections of the *I-V* curves of the PV mini-module and the electrolyser are very positive outcomes. The lessons learned are that development of device generations and, especially, finding suitable sealing materials and vendors providing these materials with high quality and short delivery time is difficult.

7.3 Links built with other deliverables, WPs and synergies created with other projects

Deliverable 3.5 was built on the outcomes of previous deliverables of WP3. The WP3 contributes to the techno-economic analysis in WP6, which links all the prototype developments achieved from WP2, WP3, and WP4 and compares the technological and economic feasibility. WP4 was also inspired by the device design which was developed in WP3. A thermally integrated PV-electrolyser prototype was provided to WP7 for outdoor demonstration. The results presented in this report can provide the

³ Kim, J. H., Hansora, D., Sharma, P., Jang, J. W., and Lee, J. S. (2019). Toward practical solar hydrogen production - an artificial photosynthetic leaf-to-farm challenge. *Chem. Soc. Rev.* 48, 1908-1971.

foundation for developing the business plan and follow-up activities and exploitation to be prepared in WP8.

7.4 Limitations of the findings

The findings show high STH efficiency and promising initial stability but are limited by possible leakage problems on longer time periods that have not been investigated so far. The efficiencies and initial stabilities are promising and are positioned among the highest reported so far for prototypes approaching 100 cm² areas. As these areas could be interconnected to form larger modules or arrays, they are of an industrially relevant area but would require more work to optimize durability and low cost for the future.

8 CONCLUSIONS AND NEXT STEPS

FeNi DLH catalysts prepared by a hydrothermal method on 100 cm² Ni foam were used as cathode and anode electrodes in a scaled-up alkaline electrolyser integrated with CIGS solar cells. The CIGS solar cell module consisted of 2×3-cells with an active total area of 82.32 cm² and had a PV module efficiency of 17.27 at 25 °C under 100 mW cm⁻² illumination. The device design consisted of a PV module / Ni plate / cathodic catalyst on Ni foam / gasket/ membrane /gasket / anodic catalyst on Ni foam / Ni plate. The maximum STH from the gas volume measurement was 13.4 % and the average STH efficiency over long operation was 11.3 % with the corresponding average hydrogen production rate of 5.74 ml min⁻¹ and area-specific hydrogen production rate of 3.74 g h⁻¹m⁻². The thermally integrated device was posted to FZJ for installation at the outdoor test site in Julich, Germany. The device was initially tested under laboratory conditions at FZJ and then installed in the outdoor test site in November 2020. Despite the “bad weather in November 2020 with a relatively low solar intensity, the first data showed that STH efficiency was in the range of 10 %. This value is still however lower than expected and indicates that something happened with the device during or after the outdoor installation. The step is to continue performing the outdoor tests to demonstrate operation on a long term and also, to assess the cause of the lower than expected outdoor performance.

9 DECLARATION BY THE DELIVERABLE LEAD BENEFICIARY

Deliverable 3.5:

- Has fully achieved its objectives and technical goals
- Has achieved most of its objectives and technical goals with relatively minor deviations
- Has failed to achieve critical objectives and/or is not at all on schedule