Optimized laser patterning for high performance Cu(In,Ga)Se₂ thin-film solar modules

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ABSTRACT

The thin-film solar cell market has seen a period of consolidation during the last years and many involved companies were forced to stop production due to increasing price pressure from competing cell technologies. Today, thin-film solar industry is gaining momentum again. Especially Cu(In,Ga)Se₂ technology evolves at high pace fired by recently achieved record efficiencies of 20.4 percent on flexible polyimide substrate [1] and 20.8 percent on glass substrate [2]. Fresh companies are preparing market entry with matured products and manufacturing technology suitable for high-volume and high-throughput production. Among these key-enabling technologies is laser patterning for cell-to-cell interconnects. Several research groups worked on efficient and reliable laser processes that are now ready for the industrial assessment. Here we present a set of work-horse processes for P1, P2 and P3 scribing of CIGS cells on glass substrate. Optimized parameters are presented for 532 nm and 1064 nm using 50 ps pulses from an all-in-fiber laser system. We further demonstrate the successful realization of functional 8-cell modules with a reduced "dead-zone" width of 70±5 µm and high efficiencies. The certified efficiency of 16.6 percent for our low-dead-zone champion module confirms the observation that shrinking of interconnects has no adverse effects on their electrical quality.

Keywords: CIGS thin film solar cell, picosecond laser, selective ablation, laser scribing, laser patterning, low dead zone, high efficiency, high throughput scribing.

1. INTRODUCTION

Today, the terrestrial Photovoltaic (PV) market is dominated by crystalline silicon wafer technology. However, thanks to extensive research and development in the field, alternative thin-film techniques are becoming more and more attractive no longer only in the esthetic architectural context but also in the cost-driven energy production market. Among the thin-film absorber materials Cu(In,Ga)Se₂ (CIGS) systems boast highest-in-class efficiency and stable performance. CIGS technology lends itself to fully integrated high volume in-line production. This is where its full potential for low production costs can be exploited. A key enabling technology for the industrialization of CIGS solar modules is laser patterning which allows the cell-to-cell interconnection to be realized with minimal loss of productive area and without the addition of foreign materials such as solder or metal conductors. These connections ("monolithic interconnects" as they are often called) consist of three (laser-)scribes which selectively remove layers of the thin-film stack between deposition processes. The cross section of the photovoltaic (PV) thin-film stack is shown in Figure 1, left side. The right side shows two typical scribe variants: the P1 scribe removes an isolating trench in the molybdenum back contact; after deposition of the CIGS absorber layer, the P2 creates a trench for locally contacting the back contact; after sputtering of the transparent conductive oxide (TCO) the P3 scribe isolates the two neighboring cells from each other by removing either only the front contact as shown in A) or by removing both, the TCO and the CIGS, as shown in B). The scribing processes are described in more detail later in the experimental part. The area consumed by the interconnect, which is no longer available for energy production, is often termed "dead zone".

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Figure 1 left: schematic overview of the PV thin-film stack. right: cross-section of a cell-to-cell interconnection illustrating how the three scribes are arranged to form the back-to-front contact. P1 stands for a scribe in the molybdenum (Mo) layer, P2 removes the CIGS film down to the molybdenum layer and the P3 scribe removes either only the transparent conducting oxide (TCO) film as shown in graphic A) or all layers down to the Mo-film, graphic B). Current flow in the interconnect is indicated by the dashed arrow. The entire interconnection zone comprising the three scribes is called "dead zone".

In earlier studies we investigated the influence of laser wavelength and pulse duration on the process quality [3–5] and module efficiency. Especially the difference between nanosecond and picosecond pulsed lasers was investigated in detail because it has considerable impact on the cost of the scribing unit in a production line. While we found nanosecond pulses to produce acceptable results for P1 and P3 in some cases, the success very much depends on the thin-film quality and the deposition technique used. In the case of the P1 process burr formation, cracking, and delamination was observed for nanosecond pulses used on high quality Mo layers with good adhesion. For the P3 process, nanosecond pulses at 532 nm can produce visually good results in some cases for interconnects according to variant A) but they are prone to creating shunts. The considerably longer interaction time compared to picosecond pulses can lead to partial transformation of the CIGS into a conductive compound [6]. P2 scribing according to variant B) failed completely when samples with good CIGS adhesion were used. Based on these findings we selected picosecond pulses with pulse duration of 30-50 ps at 532 nm or 1064 nm for our highly reliable workhorse process. In the following experimental part, each process is presented in detail with laser parameters used and process image. Further, we present achievable dead zone reduction and module performance using these processes.

2. EXPERIMENTAL

2.1 Samples

Sample coatings were grown on 50x50x1 mm³ float glass substrates at the Laboratory for Thin Films and Photovoltaics, Empa, Dübendorf, Switzerland. Molybdenum was grown by DC sputtering onto the glass substrate as electrical back contact. The Cu(In,Ga)Se₂ absorber material was deposited by co-evaporation of the elements in a low-temperature (<450°C) multistage process with NaF post deposition treatment, followed by a thin CdS buffer layer grown by chemical bath deposition and an i-ZnO/ZnO:Al bilayer grown by RF sputtering [7]. An index-matching MgF₂ layer was added to reduce reflection losses at the cell surface.

2.2 Laser sources and experimental setup

As the laser source for all experimental work in this study we used the Katana HP, manufactured by onefive GmbH, Switzerland. This is an all-in-fiber picosecond pulsed laser system which delivers <50 ps pulses at the two wavelengths 532 nm and 1064 nm. The output is a linearly polarized, single-mode Gaussian beam with a measured $M^2 < 1.1$. The maximum available pulse energy was around 11 μJ at 1064 nm and 5 μJ at 532 nm. As a special feature the laser offers an external pulse triggerable operation mode. This means the laser can, as an example, operate in "slave-mode" together with a machine and send pulses as a response to the encoder signal of a machine axis. The optical beam path in scribing experiments was kept as simple as possible with a plano-convex focusing lens as the only refractive element.

Figure 2 Flexible lab scribing tool for process development at BUAS-ALPS lab. Working area: $340 \times 750 \text{ mm}^2$ with micrometer precision at up to 2 m/s. An optical experimentation platform on top and the two working heads allow to quickly switch between different laser systems and wavelengths and also offer space for extensive manipulation of the beam. The right photograph shows the standard Katana all-in-fiber picosecond pulsed laser system manufactured by onefive GmbH. The laser is currently being released in a new version with higher performance.

All scribing experiments and functional modules were made on a computer controlled lab scribing tool built by Solneva SA, depicted in Figure 2. The machine is equipped with industrial-grade linear motors that guarantee an accurate and fast sample positioning and stable performance. The working area measures $340 \times 750 \text{ mm}^2$ and the sample can be moved with micrometer precision at speeds up to 2 m/s. A slower third axis moves the focusing element. An experimentation platform on top of the scribing machine allows the installation of various types of laser sources and provides enough space for manipulation of the beam before it is guided to one of the two working heads. The setup is optimized for maximum flexibility and allows to switch between different working laser sources and wavelengths in a matter of seconds. The control software can process scripts for automated parameter studies including variation of laser parameters, motion profiles and focal distance. Finally, a machine vision system can be used for direct inspection of the scribe and surface quality and for sample alignment.

2.3 P1 scribe

At the production stage of the P1 process, a glass substrate with a molybdenum (Mo) coating on one side has to be patterned. The 500 to 750 nm thick Mo layer must be sub-divided into electrically isolated lanes which are the base for the later individual PV cell strips. Based on our earlier experience[3] and other published results[8–10] we have chosen the substrate scribing configuration where the molybdenum layer is illuminated from the backside through the glass substrate. The rapid expansion of the Mo at the interface creates a thermo-elastic shock wave, which separates and ejects the Mo chip from the substrate. Quality criteria for a good P1 scribe are minimum burr formation, minimum heat deposition at the scribe border, no delamination and no cracking of the Mo layer and no damage of the substrate. While these requirements can be satisfied relatively easy for sample material with mediocre adhesion of the Mo film it becomes quite challenging to meet all requirements when samples with a high-quality back contact are to be structured. We found it to be advantageous to use short picosecond pulses under these circumstances. We could demonstrate that a reduction of the pulse length to 10-50 ps can solve most of the problems and greatly expand the process window with respect to variation of focal position and fluence.

Figure 3 Typical process image for the P1 scribe achieved with 50 ps pulses at 532 nm, very similar results are observed for 1064 nm. White spots in the scribe are no substrate damage but a surface charging effect observed during SEM analysis.

As Figure 3 demonstrates, we can achieve a very good P1 scribe quality also on high quality molybdenum films using 50 ps pulses. The scribe borders are characterized by a very clean, burr-free rupture of the molybdenum layer, no melting at all and no delamination. Cracks rarely occur and if they do, they follow the shape of the ablated spot and never run radially into the film. An indication of how low the impact of the scribing is on the integrity of the Mo film gives the detail picture of a scribe crossing on the right side in Figure 3. The process works as usual even when it hits a region, which has been irradiated by the laser before.

Table 1 Typical scribing parameters used for P1 processes at the two wavelengths (50 ps pulse duration):

wavelength	532 nm	1064 nm
repetition rate	50 kHz	50 kHz
overlap (velocity)	40% (600 mm/s)	20% (1000 mm/s)
pulse energy	4.3 μJ	9.5 μJ
spot radius w_0	10.0 μm	12.5 μm
fluence	2.7 J/cm^2	3.9 J/cm^2

2.4 P2 scribe

During the P2 process step an electrical contact has to be prepared for the later TCO front contact. This can happen either by selectively removing the CIGS film along the scribe or by locally transforming it into an electrical conductor[11]. We investigated several different processes in the past for producing functional modules and at the end of every series of experiments it turned out, that using low energy picosecond pulses at high overlap for the P2 process produced the best performing modules. This is in accordance with results of other groups[9,12]. It became clear that this should be our workhorse process as it is also robust, reliable and controllable. Besides the good electrical performance of the contacts it produces smooth scribe borders and leaves the underlying Mo back contact intact. The fact, that picosecond pulses are used helps to keep the heat-affected zone along the scribe small. The process works at 532 nm and 1064 nm wavelength and as Figure 4 shows, both wavelengths produce scribes with similar characteristics: the Mo back contact is not damaged and the scribe borders are smooth with some melting. Despite this similarity, the ablation mechanism is not the same. While most of the green light is absorbed at the surface, the penetration depth in the CIGS is much bigger for the NIR. A detailed description of the P2 process at 532 nm can be found in our earlier work[3,4], the description of the P2 process in the NIR is included in [12].

Figure 4 Process image (SEM) of the P2 scribing process at 532 nm (left) and at 1064 nm (right).

Table 2 Typical scribing parameters used for P2 processes in the green and NIR (50 ps pulse duration):

wavelength	532 nm	1064 nm
repetition rate	200 kHz	50 kHz
overlap (velocity)	99% (50 mm/s)	96% (50 mm/s)
pulse energy	2.0 µJ	12 µJ
spot radius w_0	12.5 µm	12.5 µm
fluence	0.81 J/cm ²	4.9 J/cm ²

2.5 P3 scribe

The third scribing process electrically isolates the individual PV cells by removing the TCO front contact layer locally. This process step completes the serial interconnection of the cells. As shown in Figure 1, there are two possibilities to achieve that – by removing the TCO only or by removing all layers down to the Mo back-contact. Both strategies can be equally successful. The main problem here is CIGS melting, but – other than in the P2 process – we cannot tolerate electrically conducting zones in the CIGS as these would create shunts that directly affect module performance. On our samples the TCO used as front contact is (ZnO:Al). This material has by design a high transmission in the near infrared (NIR) to the visible spectral range but the absorption is high in the UV. Direct ablation of the TCO layer has been reported using picosecond pulses at 266 nm and 355 nm[13,14] or femtosecond pulses[15]. However, we did not like the idea of changing the laser wavelength for the P3 process and worked on P3 scribing processes at 532 nm and 1064 nm as described in[5,12,16,17]. Due to the different penetration depth of the two wavelengths in the CIGS the two processes work differently and lead to different process images. At 532 nm, the major part of the pulse energy is absorbed at the interface between TCO and CIGS. The shockwave separates the TCO layer from the CIGS and ejects the TCO fragments. In this process, the CIGS layer is only used as an absorber for the induced lift-off process, the CIGS itself stays in place. The process image is shown in Figure 5. The scribe border shows a clean rupture of the TCO and some signs of melting, although concentrated mainly in the middle of the scribe. Nevertheless, this melt potentially creates “horizontal” shunts. While the lift-off process works perfectly also with short nanosecond pulses, we prefer to exclude potential problems with shunts by using ultra-short pulses. The P3 process at 1064 nm has been described in literature [14]. The pulse energy is here absorbed in a much larger volume in the CIGS due to the larger penetration depth in the NIR. The shockwave created is not strong enough to lift-off the TCO chip at once. The process image shown in Figure 6 is therefore very similar to the P2, only that here the width of the exposed back contact is not relevant. The melt along the scribe border tends to create “vertical” shunts which can be avoided almost completely by using ultrashort pulses such as the 50 ps used here.

Figure 5 Induced lift-off P3 process at 532 nm, 50 ps.
The CIGS, which acted as an absorber remains in place and the TCO chip is ejected.

Figure 6 P3 process at 1064 nm, 50 ps. Process image similar to the P2 scribe: CIGS is partially removed, melt is produced along the scribe borders..

Table 3 Optimized process parameters for the P3 scribe at 532 nm and 1064 nm:

wavelength	532 nm	1064 nm
repetition rate	50 kHz	50 kHz
overlap (velocity)	30% (1200 mm/s)	92% (100 mm/s)
pulse energy	3.0 μJ	12 μJ
spot radius w_0	15 μm	12.5 μm
fluence	0.85 J/cm^2	4.9 J/cm^2

3. FUNCTIONAL MINI MODULES

We produced several series of 8-cell mini modules. Each series comprised 8 to 16 mini-modules structured with different laser scribing parameter combinations according to the pattern in Figure 7. Finally, a series of four functional-modules with selected optimized parameters was manufactured with reduced dead-zone width.

Figure 7 Scribe pattern for functional mini modules. The module consists of eight individual cells with an active area of $4 \times 40 \text{ mm}^2$ each. The substrate dimensions are $50 \times 50 \text{ mm}^2$. The marks in the corners are used for fine re-positioning of the sample between process steps. The isolation scribe is described in [3].

3.1 Dead zone reduction

We tested the feasibility of low dead-zone modules and how dead-zone reduction influences module performance. The first concern was that laser illumination could affect physical properties of the film in the neighborhood of a scribe in a way that process mechanics of the following scribe are changed significantly. Second, we wanted to find out if electrical quality of the interconnect suffers when scribes are placed very close to each other. This concern is aimed especially at the P3 which deposits large amounts of energy very close to the critical P2 contact zone. We could demonstrate, that we can mechanically and optically handle an inter-scribe distance of 15-30 μm center-to-center on a 50 x 50 mm² sample. We achieved a reduced dead-zone of 70 $\pm$ 5 μm on several entire functional 8-cell modules. We could further demonstrate that this reduced dead-zone has no adverse effect on the electrical quality of the interconnect and the module performance. The increase in active cell area directly increased overall electrical yield and the modules with the lowest dead zone width were also the ones with the best module performance.

3.2 Electrical analysis

The functional solar modules were analyzed by means of illuminated current voltage (JV) characteristics at the Empa laboratory in Dübendorf, Switzerland. The JV measurements were accomplished according to the international standard IEC 60904-1 under standard test conditions with AM1.5G irradiation spectrum and 1000 Wm⁻² irradiation intensity. The devices under test were kept at constant temperature of 25°C during measurement. The area of the devices was determined from a 1200 dpi scan using graphical software. Typically, the active surface was around 12.6 cm² per module and 1.56-1.59 cm² per cell (8 cells). We prepared two varieties of functional modules patterned at different laser wavelength; one group had P2 and P3 structured at 532 nm the other group at 1064 nm. Scribing parameters were set according to the tables presented before in section 2. Measured values of the two best performing low-dead-zone modules for each wavelength are given in Table 4.

Table 4 JV characteristics for two representative modules. The module in the left column was structured at 532 nm, 50 ps, the module in the right column at 1064 nm, 50 ps. Electrical parameters of the modules: V_{oc} : open circuit voltage, J_{sc} : short circuit current, FF: fill factor, η : efficiency (designated illumination area).

	module 1 532 nm	module 2 1064 nm
V_{oc}/cell V	707	665
J_{sc} A/c	31.7	33.5
	74.5	65.3
η	16.7	14.5

The measured J_{sc} for the module 1 structured at 1064 nm is higher than for module 2, whereas module 1 dominates all other categories. Module 1 shows an excellent open circuit voltage of 707 mV, which is an indication for a high parallel resistance and good electrical isolation of P1 and P3 scribe. Module 2 has a good J_{sc} indicating a good electrical contact between TCO and Mo in the P2 scribe. This is a trend, which was regularly observed but not confirmed yet due to insufficient statistical data. Assuming the two modules have comparable quality thin films we could say 532 nm produce better P3 results and 1064 nm better P2 results. For the latter we did not find any evidence in the literature. For the P3 process on the other hand a degradation of the solar cell performance was observed for P3 scribing at 1064 nm [16,18]. When the necessary energy input for a P3 scribe at 532 nm and 1064 nm is compared (see Table 3), it becomes clear, that the amount of energy needed in the case of 1064 nm is much bigger than for 532 nm and therefore also all adverse side effects such as melting and material modification at the scribe border get stronger (compare Figure 5 to Figure 6). Our champion module 1 was later independently certified with a confirmed efficiency of 16.6 percent module efficiency (designated illumination area).

4. CONCLUSIONS AND OUTLOOK

In this study we have shown that stable and reliable scribing processes exist for structuring of Cu(In,Ga)Se₂ solar modules on glass substrate with ZnO/ZnO:Al front contact. We presented an entire set of process parameters for P1, P2 and P3 using 50 ps pulses from an all-in-fiber laser system at 532 nm or 1064 nm. We demonstrated the feasibility of reduced dead-zone functional modules with a dead-zone width of only 70±5µm. The reduced size of the interconnects and their excellent electrical quality directly translates into better module performance. This is underlined by the certified module efficiency of 16.6% achieved with a low-dead-zone module.

Now that reliable laser scribing processes and laser sources are available, these results need to be transferred into industrial production. In the process of industrialization, the focus shifts from understanding of the process mechanics to more practical aspects of laser processing such as long-term performance and stability of the laser system, beam delivery and machine integration of the laser device and of course total cost of ownership. Research institutes should accompany this transfer process from lab to production line and work together with the early adopters of all-laser-scribing among the solar module producers. An initiative aiming in this direction is the recently started European FP7 project APPOLO (www.appolo-fp7.eu). In this project comprising 21 partners from all over Europe, a hub of five application labs will develop and establish, together with end-users and suppliers, assessment procedures for laser equipment and laser processes used in industrial production. One of the main fields of activity is the assessment of scribing processes and necessary laser equipment for high-volume, high-throughput solar cell production.

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