Diffractive Multiplexing for High-Throughput Roll-to-Roll Laser Patterning of Flexible Organic Photovoltaic Modules

Cooperative Research and Development Final Report

CRADA Number: CRD-17-00721

NREL Technical Contact: Bertrand Tremolet de Villers

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Office of Energy Efficiency & Renewable Energy
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Contract No. DE-AC36-08GO28308

Technical Report
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NOTICE

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Cooperative Research and Development Final Report

Report Date: September 6, 2023

In accordance with requirements set forth in the terms of the CRADA agreement, this document is the CRADA final report, including a list of subject inventions, to be forwarded to the DOE Office of Scientific and Technical Information as part of the commitment to the public to demonstrate results of federally funded research.

Parties to the Agreement: SolarWindow Technologies, Inc.

CRADA Number: CRD-17-00721

CRADA Title: Diffractive Multiplexing for High-Throughput Roll-to-Roll Laser Patterning of Flexible Organic Photovoltaic Modules

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Sponsoring DOE Program Office(s): Office of Energy Efficiency and Renewable Energy (EERE), Advanced Manufacturing Office (AMO)

Joint Work Statement Funding Table showing DOE commitment:

Estimated Costs	NREL Shared Resources a/k/a Government In-Kind
Year 1	$145,000.00
Year 2, Modification #1	$145,000.00
Year 3, Modification #2	$.00
Year 4, Modification #3	$.00
TOTALS	$290,000.00

Executive Summary of CRADA Work:

The purpose of this project is to demonstrate a cost-effective, high-throughput roll-to-roll (R2R) process for patterning of flexible, semitransparent organic photovoltaic (OPV) modules by developing diffractive optics-based laser multiplexing (DOL Multiplexing). DOL Multiplexing allows a single, high-powered laser source to perform parallel scribing across the R2R web width in a manner compatible with high process speeds. Such a process could have enormous benefits in terms of increased process speeds and reduced costs, both up-front capital costs, and long-term operational costs, over galvanometer-based step and scan methods or many-laser systems.

CRADA benefit to DOE, participant, and US Taxpayer:

- Assists laboratory in achieving programmatic scope,
- Enhances the laboratory’s core competencies,
- Uses the laboratory’s core competencies.

Summary of Research Results:

The scope of this project was organized into five stages, as described in **Table 1** below. Stage 1 was to design and build the diffractive optic laser multiplexed scribing station and integrate it into the R2R webline. Part of Stage 1 was to use a sheet-to-sheet single-laser scribing unit as the baseline for comparison with the multiplex system. Data from the single laser scribes were used to design the multiplex scribing system. Stage 2 was to demonstrate feasibility of the multiplexed scribing. Stage 3 was to compare functionality of the multiplexed scribed samples to single beam scribed control samples. Stage 4 was to achieve entirely multiplexed scribed (P1, P2, and P3 steps) samples in a fully processed module. Stage 5 was to gradually increase the process speed to demonstrate suitability of the multiplexed scribing process for true high-throughput R2R manufacturing.

Table 1. Proposed project workplan proposed in five stages with subtasks for each stage.

Stage	Task
1	Select Consortium R2R Line
1	Design DOL Multiplexed Scribing Station
1	Select/Design DOE
1	Fabricate DOL Multiplexed Scribing Station
1	Laser Beam Alignment and Tuning
2	Finalize R2R Web Substrate
2	Optimize R2R Coated Films
2	Optimize Single-Beam Scribing: P1, P2, & P3
2	Tune DOL Multiplexed Laser Output
2	Perform Slow DOL Multiplexed Scribing
2	**Tune Process for Crossweb Uniformity**
GNG	**Uniform Web Width Scribing?**
3	Prepare All Single-Scribe Control MM
3	Prepare Single DOL Multiplex-Scribed MM
3	Compare MM Performance and Imaging
4	Prepare new Optimized Coatings
4	Perform DOL-Multiplexed Scribing: P1, P2, & P3
4	**Prepare, Test, and Image MM Samples**
5	Increase DOL Multiplexed Scribing Speed
5	Analyze Scribe Quality, Uniformity & Stability
5	MM Performance as Function of Process Speed
GNG	**Performance Scalable to High-Throughput?**

A summary of the research results for each stage is presented as follows:

Stage 1: design and build DOL Multiplexed Scribing Station and integrate into R2R line.

Figure 1: The NREL "metrology" roll-to-roll was selected as the appropriate tool for integration of the multiplexed laser ablation web processing.

The concept for this project was to use a single laser source and multiplex the laser beam into multiple beams or lines so that all P1s, then all P2, and all P3 scribes could be made individually and simultaneously. The resulting multiplex system would be incorporated onto a R2R webline (Stage 1, Task 1), and the NREL "metrology" R2R was selected, as shown in **Figure 1**. A laser safety enclosure/DOE scribing station (Stage 1, Tasks 2 and 4) was designed and constructed around the R2R to house the DOE optics and contain the full laser beam path. Custom DOE optics were fabricated using an external specialty company (Stage 1, Task 3). After EHSQ approval to operate the laser system, beam alignment and tuning was successfully completed (Stage 1, Task 5)

Stage 2: demonstrate feasibility of the DOL Multiplexed Scribing.

Due to cost, availability, and appropriate mechanical and optical properties, polyethylene terephthalate (PET) was selected as the most compatible web substrate for the project (Stage 2, Task 1). On NREL's deposition R2R, approx. 6 meters of OPV material with several layers was fabricated. The first coating was transparent electrode (12in wide) on a PET web. The photoactive and charge-selective layers of our solar cell stack (6in wide) were subsequently coated. The result was about 2 meters of each combination of layers—layer 1 for P1 scribing & layers 1+2+3 for P2 scribing; layers 1+2 for diagnostic purposes (Stage 2, Task 2).

Using our single-scribe laser system, the P1 laser scribe was explored. In this system, the laser is fixed and the sample platform moves. The laser power and the platform speed were adjusted to provide a range of power densities. Promising initial results provided a sizeable "processing window" that yielded electrically-isolated P1 scribes; varying from low power with slow speeds to higher power with fast speeds. Similar to the P1 scribe optimization, processing windows for P2 and P3 laser scribes were identified (Stage 2, Task 3).

Our first series of laser scribing was done with a "slow" web speed of 2ft/min. With only gross alignment of our optics, 14 isolation lines into the transparent conducting film was successfully scribed (Stage 2, Tasks 4 and 5). **Figure 2** shows optical microscope images of the scribes. These P1 scribes must electrically isolate the bottom electrodes of each photovoltaic cell from its adjacent cell. The widths of the laser scribes vary from approx. 28 to 47microns, however they each still successfully isolated 13 cells. Although not necessary for PV module performance, fine tuning of the optics and laser powers to achieve more uniform scribing widths was performed to help with final sample aesthetics (Stage 2, Task 6, and GNG milestone completed).

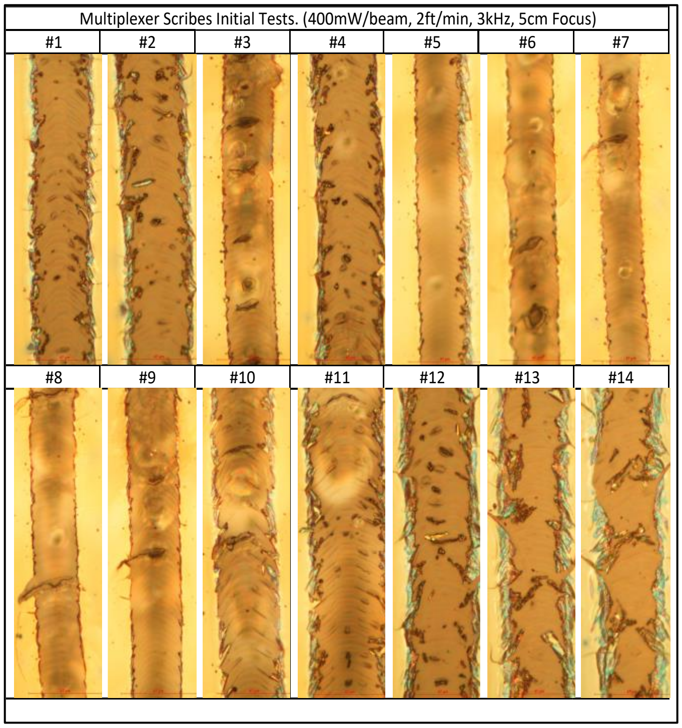

Figure 2: Optical microscope images of 14 laser scribes in transparent conducting film on plastic substrates.

<u>Stage 3</u>: compare functionality of DOL Multiplexed Scribed samples to single-beam scribed control samples.

NREL scribed several meters of P1 scribes on transparent conducting film on plastic substrate on their R2R laser scribing system (Stage 3, Task 2). Comparable samples were made with single-laser P1 scribes (Stage 3, Task1). Approximately 5-in x 4-in sections were cut and coated with photoactive ink and finished with hole-collection and top contact layers. **Figure 3** shows an example of completed mini-modules. None of the mini-modules had good performance because most were shorted. Using the un-scribed portions of our transparent conducting film on plastic substrates, small-area solar cells were fabricated with various combinations of different photoactive inks (blade-coated) and evaporated the top contacts. None of the devices worked well. The overall conclusion from these efforts was that the electron charge-selective layer (or ESL) was the culprit for the poorly-performing devices. To address the issue, a new ESL precursor solution chemistry was developed.

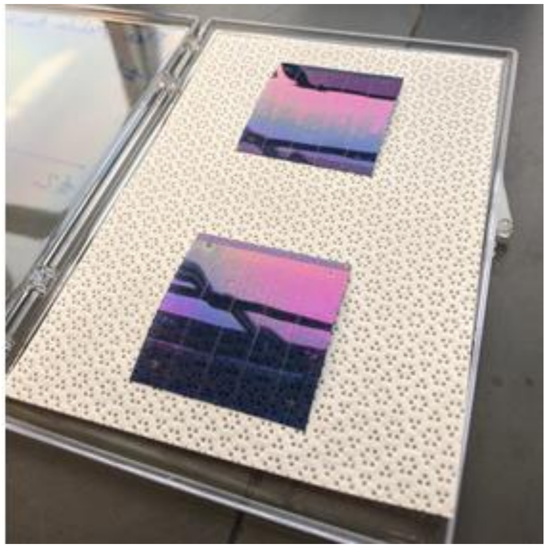

Figure 3: Example of PV mini-modules consisting of three cells.

Diagnosis of module interconnect failures using a combination of optical microscopy, film surface profiling, and dark lock-in thermography (DLIT) (**Figure 4**) identified problems with 1) chemistry-related morphology of the ESL and 2) bottom contact delamination during P1 laser scribing (Stage3, Task 3).

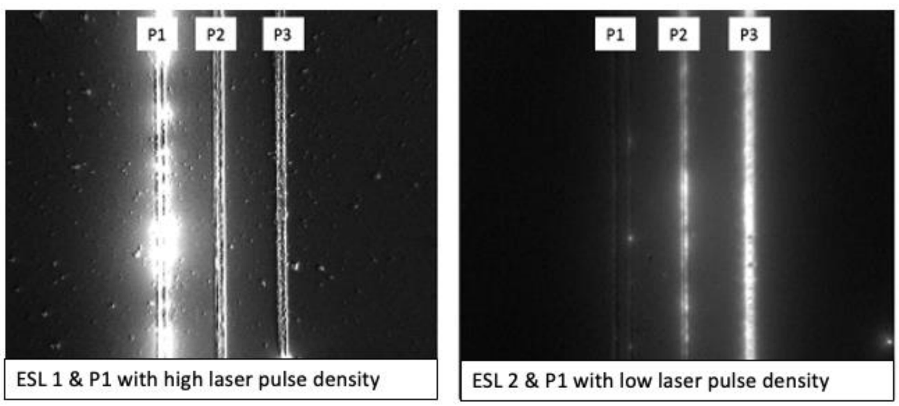

Figure 4: DLIT images of flexible organic PV mini-modules with different electron charge-selective layers (ESL) and two different P1 laser scribing settings. Left image: Bright current spots caused by the electrodes short circuiting indicate failed P1 scribe. Right image: With different ESL and laser scribe parameters, problems with P1 were eliminated.

<u>Stage 4</u>: achieve entirely multiplexed scribed (P1, P2, and P3 steps) samples in a fully processed module.

Although the scribing processes with the flexible plastics substrates was significant improved, and issues with unoptimized ESL coating were resolved (Stage 4, Task 1 and Figure 4), a decision was made to use flexible glass as an alternative to the PET. The glass has many advantages, specifically better optical uniformity, much smoother surfaces, and most critically, better adhesion with the transparent conducting electrode deposited on top of it. Integration of the flexible glass onto the R2R multiplexed laser scribing tool was achieved. **Figure 5** shows the surface profiles of initial laser P1 scribes using the R2R multiplexed laser. P1 scribes with minimal sidewalls were fabricated by tuning the repetition rate of the laser pulses striking the ITO/glass, for example the blue curve shows sidewalls less than 60 nm.

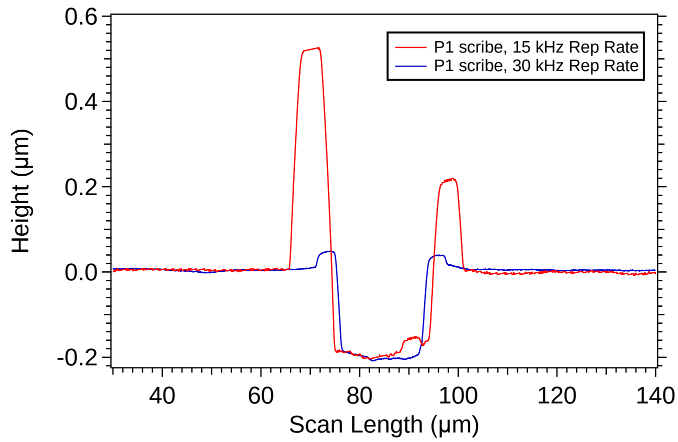

Figure 5: Surface profile scans across laser P1 scribes of ITO/glass showing that doubling the laser pulse repetition rate resulted in significantly improved edge features.

Having overcome the major problems with P1 scribes, the R2R multiplexed laser tool was successfully used to scribe P2 lines, and eventually to finish a working PV module comprising 12 individual sub-cells connected in series through the P1/P2/P3 interconnects (Stage 4, Tasks 2 and 3 and **Figure 6b**). **Figure 6a** shows the increase in voltage that results from increasing the module size from 1 to 12 cells (black curve). However, each cell does not contribute an equal amount of voltage (grey curve), suggesting that some of the cells are suffering performance losses. The fraction of the open circuit voltage (Voc) contributed by each individual cell is included in the figure, for example, cell #1 contributed ~13% whereas cell #3 only contributed ~4% to the module's Voc.

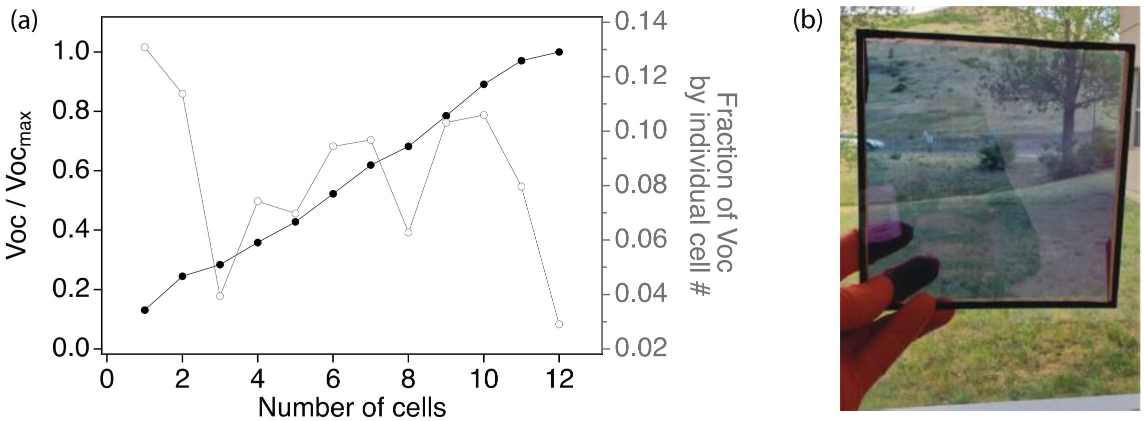

Figure 6: a) Increase in the PV module size from 1 cell to 12 cells produced an increase of the module's Voc. (b) A photo of a completed semitransparent PV module on flexible glass.

<u>Stage 5</u>: gradually increase the process speed to demonstrate suitability of the multiplexed scribing process for true high-throughput R2R manufacturing.

The project was interrupted by the COVID-19 pandemic and the laboratory was closed for a significant amount of time. As a result, the project schedule was delayed. Stage 5 was not completed. However, the project was deemed successful having completed Stages 1-4. Working full modules were fabricated on flexible glass using the single line laser scriber as a comparison to R2R-processed modules. Module performance was similar between the two scribing processes and demonstrated the success of the multiplexed laser scribing on the R2R. The fastest R2R scribing obtained was 14 simultaneous scribe lines at a rate of 2 ft/min. Although not demonstrated in this project, it has been possible to increase this speed by at least 2x.

<u>Subject Inventions Listing</u>:

None

<u>ROI #</u>:

NREL ROI-20-67 (Inactivated), *Diffractive Multiplexing and Beam Shaping for High-Throughput Roll-to-Roll Laser Patterning of Photovoltaic Modules*. Tremolet de Villers, Bertrand; van Hest, Marinus (Maikel); Heinemann, Matthew; Whitaker, James. No patent application was filed.