Article

Printability of Collecting Electrode Using AJP for New Construction of Photovoltaic Device

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Abstract: In 2018, the European Parliament and Council laid down a directive about the promotion of the use of energy from renewable sources connected with the Paris Agreement, which sets a global ambition on climate change mitigation through deep and fast cuts in greenhouse gas emissions. Since then, the science world has been even more focused on the development of green technologies such as wind farms, waterpower stations, and photovoltaics as the European Union is preparing to shift to renewables-based energy systems. Each green power technology has its own problems and limitations. Nevertheless, for environmental protection, new power technologies have to be implemented in the near future as primary power sources. Described in this article is the application of aerosol jet printing in manufacture of photovoltaic cells, moving the technology boundaries further toward highly efficient, cost-effective, green power production. The research focused on utilizing aerosol jet printing technology to create finger-shaped collecting electrodes on a newly constructed, non-silicon photovoltaic cell, based on metal oxides. Three commercial nanosilver inks were investigated considering their printing parameters, printability on the specified substrate (AZO-coated glass, AZO-coated copper plate), resistivity of the cured composite, quality of the overprints, and application in photovoltaics. As a result, we obtained finger-shaped collecting electrodes with a resistivity of 3.5 µΩ·cm and 8 µm width, which compares well with the literature.

Keywords: aerosol jet printing; non-silicon photovoltaics; collecting electrode

1. Introduction

In parallel with economic and technological development, global needs for electricity have risen. This phenomenon is caused by, e.g., progressing electrification or production lines automatization. Existing power plants are restructured to improve their efficiency and new ones are constantly being built. At the same time, requirements for power engineering are evolving. These factors drive the research for new, effective, green methods of obtaining electrical energy [1–12]. In 2015, energy obtained from renewable energy sources consisted of 70% water energy and 15% wind energy, with solar energy contributing only 5% [13]. Present data show that solar energy technologies need to be expanded and developed. Gerhard Knies claims that the earth’s desert areas receive 8 × 107 TWh of solar energy annually [14]. The Energy Information Administration (EIA), the American government agency collecting information related to the usage of electricity around the world, stated that in 2015 humanity generated 2.3 × 104 TWh [13]. These numbers show colossal potential of solar power plants and the promising direction of power engineering.
The above data provide the motivation to develop solar energy conversion technologies to both improve the efficiency of photovoltaic devices and to reduce their production costs.

An idea to decreasing photovoltaic devices’ production costs is the utilization of additive manufacturing technologies such as aerosol jet printing. Considering the importance of the shape and quality of the collecting electrode \([15–17]\) in terms of PV cell efficiency and fill factor, this technology can provide a number of advantages in the photovoltaic industry:

- Capability of printing narrow overprints (10 \(\mu m\) or narrower);
- Low resistivity of printed paths;
- Good adhesion to the substrate;
- High-speed printing, considering width of overprint.

Throughout the world, scientists are researching how different approaches to creating top collecting electrodes using additive manufacturing technologies impact a PV device’s efficiency and fill factor \([15–23]\). As grid-shaped collecting electrodes placed on the top of photovoltaic cells should be as narrow as possible, maintaining high conductivity, aerosol jet printing and other additive manufacturing technologies are competing with traditional techniques for manufacturing these electrodes.

Table 1 presents recent research related to the usage of additive manufacturing technologies in photovoltaics, showing aspects in which AJP may improve parameters such as the fill factor (FF) or power conversion efficiency (PCE). As the requirements for collecting electrodes face the problem of active layers with high resistance, it is desirable that the resistance be lowered by creating dense and narrow conducting lines on the surface of the photovoltaic device. This can be achieved with small line width, allowing the application of smaller pitch sizes and at the same time reducing shadowing of the photovoltaic cell.

| Reference | PV Type | Active Material | Printing Method | Electrode Type | FF (%) | PCE (%) | Line Width [\(\mu m\)] | Pitch Size [mm] | Shadowing [%] |
|-----------|---------|----------------|----------------|----------------|--------|---------|-------------------|---------------|--------------|
| [15]      | Organic | P3HT:PCBM (10:9) | AJP            | 2D grid       | 54.6   | 1.64    | 30                | 0.8           | 7.5          |
| [16]      | Organic | P3HT:PCBM (10:7) | AJP            | 1D grid       | 56     | 2.64    | 70                | 1             | 7            |
| [17]      | Organic | P3HT:PCBM (1:1)  | IJP            | 1D grid       | 53     | 2.34    | 200               | 2             | 10           |
| [18]      | Organic | P3HT:PCBM (1:1)  | IJP            | 2D grid       | 50.2   | 2.47    | 1000              | 20            | 10           |

It can be seen that electrodes printed with AJP offer a reduction in the shadowing level to 3.75%. Compared to shadowing levels common in the photovoltaic industry (10%), this is a reduction of over 60% \([24]\). However, modification of line width and pitch size may lead to a rise in FF and PCE.

The research described in this paper was conducted to identify possibilities related to AJP application in the PV industry. It shows that fundamental parameters of the collecting electrode, such as high conductivity or narrow, fine geometry, can be achieved using this method. Incorporating AJP into the PV industry can improve PV device work parameters through the manufacture of cheaper and better quality electrodes.

The PV examples listed in Table 1 were annealed at a temperature of 120–200 °C, but there was no conclusion regarding the impact of this on the condition of high-temperature-sensitive materials. As our aim was to design a manufacturing process with consideration to this sensitivity, we conducted measurements of the resistance changes under different curing conditions.

2. Materials and Methods

Inks used in this study were delivered by UTDots (Ag25X, Ag40X, 2716 Clark Rd Suite E, Champaign, IL 61822, USA) and Clariant (Prelect TPS 50G2). Their viscosity was measured with the LVDV2T Viscometer from Brookfield.

In the research described, an Optomec Aerosol Jet Printer with a 100 \(\mu m\) nozzle was used to create overprints on AZO-coated substrates (glass and copper plates), which
simulated the surface of a PV device (Figure 1). Atomization of inks was conducted with a pneumatic atomizer (PA) and an ultrasonic atomizer with half-spherical bottle base (UA), which are part of the Optomec machine. Additionally, a UA created and described in a previous publication [25] was used. This custom atomizer can move the ink bottle in up–down and forward–backward directions, as well as rotating the bottle in the plane constructed with those axes. Overprints were cured in a laboratory furnace at 200 °C for 1 h. Conductivity of printed samples was measured with a Keysight 34461A multimeter. The height and width were measured with a Vecco profilometer.

Figure 1. Schematic diagram of the new photovoltaic device: 1, Al/Cu substrate; 2, CuO/SnS layer; 3, ZnO layer; 4, AZO layer; 5, printed finger electrode; 6, smartwire.

3. Results
3.1. AZO Thermal Curing

The construction pictured in Figure 1 consists of materials, of which one is vulnerable to high temperature: aluminum-doped zinc oxide (AZO). Therefore, before introducing specific thermal curing conditions for overprints as described in the research, it was necessary to check the influence of curing temperature and curing time on AZO electrical properties to protect the device from damage caused by thermal degradation. Deneault et al. undertook a sintering study for AJP conductive inks. UTDots Ag40X showed a dramatic increase in conductivity when the curing temperature was increased from 185 to 205 °C from $7 \times 10^5$ to $10^6$ S/cm. Further elevation of temperature improved the conductivity. Thermal curing below 185 °C yielded non-conducting paths. Similar results were obtained for Clariant Prelect TPS50 ink, but its conductivity above 205 °C in the sintering process was $2 \times 10^6$ S/cm [26], leading to the assumption that 200 °C is the minimal acceptable curing temperature.

To select the optimal thermal treatment, a series of resistance measurements were made at various temperatures and times of sample drying. The results are presented in Figure 2 and in Table 2. To qualitatively evaluate the resistance, samples of AZO-coated glass 25 mm × 25 mm were prepared and sections of 10 mm length were marked on each to enable measurements to be repeated in the same area.
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![Figure 2. Changes in AZO resistance with different curing parameters.](image)

Regarding differences between pre-cured and post-cured samples resistance, Table 2 shows percentage changes. This experiment demonstrated that temperatures above 225 °C cause serious changes in electrical properties with even a 0.5 h curing process. Slight resistance fluctuations of ±3% during measurements were considered to be measurement error. A 200 °C 1 h curing process was considered optimal for maintaining AZO properties, while being sufficient for drying overprints. Further, it does not change the resistance of the AZO layer by more than 5%.

Table 2. Changes in resistance of samples cured for 0.5 h and 1 h at different temperatures, expressed as a percentage.

| Curing Temperature (°C) | Curing Time |
|-------------------------|-------------|
|                         | 0.5 h       | 1 h         |
| 200                     | −1.75%      | 2.60%       |
| 225                     | −2.83%      | 10.07%      |
| 250                     | 31.49%      | 79.39%      |
| 275                     | 77.03%      | 154.72%     |
| 300                     | 355.70%     | 758.11%     |

3.2. Setting Printing Parameters

Depending on the type of ink, specific methods of atomization were used. For Prelect TPS 50, the manufacturer recommends using a PA but the literature shows that it can be atomized ultrasonically with satisfactory results [27,28]. The UA method is a simple system with two types of gas flow: atomization flow and sheath flow. The PA system is more complex and has three types of gas flow, the additional being exhaust flow. While the PA can be used with inks of wider viscosity range, up to 1000 cP, the scope of the UA is limited to 1–5 cP. It was concluded that it is easier to achieve solid atomization with a UA, as encountered problems can be diagnosed and solved promptly, although it is not suitable for every material. The PA is more versatile but difficulties are often more complicated and solutions are time-consuming, hindering satisfactory results being obtained. Overprints were made using the PA and UA. The custom UA did not provide any atomization for pure TPS 50, nor when diluted in water 1:1 v/v. For UTDots inks, the manufacturer recommends a UA. The PA was also tested with unsatisfactory results. The custom UA produced the best atomization. The position of the ink bottle was adjusted experimentally for each printing process to achieve the same level of atomization. During works described in
Table 3. Established printing parameters for inks used: AF, atomization flow; ExhF, exhaust flow; SF, sheath flow. Considering PA physics, formula \([\text{ExhF} - \text{AF}]\) gives the actual value of the atomization flow at the nozzle.

| Ink            | Atomization Type | AF (sccm) | ExhF (sccm) | SF (sccm) | Velocity (mm/s) | Nozzle Size | Plate Temp. (°C) | Line Width (µm) |
|----------------|------------------|-----------|-------------|-----------|-----------------|-------------|------------------|-----------------|
| Clariant TPS50 | Pneumatic        | 900       | 867–875     | 50        | 1               | 100         | 60               | 8–30            |
| Clariant TPS 50| Ultrasonic       | 22–25     | -           | 22–26     | 0.5–1           | 100         | 60               | 25–50           |
| UTDots Ag25X  | Ultrasonic       | 16–18     | -           | 20–33     | 1               | 100         | 60               | 20–50           |
| UTDots Ag40X  | Ultrasonic       | 11–18     | -           | 30–50     | 1               | 100         | 60               | 2–30            |

Figure 3. Examples of well-printed lines: (a) Clariant Prelect TPS50 (13 µm); (b) UTDots Ag25X (34 µm); (c) UTDots Ag40X (33 µm).

3.3. Resistivity Calculations

To understand the electrical properties of the inks used, measurements of the overprints’ resistance and cross-sections were made. The average cross-sectional area was calculated using information from three different longitudinal coordinates. As the obtained cross-sections were triangular-shaped, the area was approximated using the triangle area formula (Figure 4).

Figure 4. Examples of calculating cross-sectional area for samples printed with Clariant Prelect TPS50. Peaks outside of the triangles represent overspray. Cross-section at the beginning (a) and at the end (b) of the line.

The width of samples was also inspected with a digital microscope to confirm the values, considering the uncertainties of the measuring instruments. The profilometer measured the width at certain cross-sections, making this method sensitive to any irregularities (Figure 5b). Figure 5b shows the detailed characteristic shape of lines printed using the AJP method (overprint core of 10–15 µm and overspray alongside the line).
Figure 5. Different methods of measurement compensate for irregularities or show them (Clariant Prelect TPS50). (a) optical microscopic picture and (b) profilometer scan.

The calculated resistivities for each sample of each ink were used to create box-and-whisker plots (Figure 6). All gathered data gave the final values shown in Table 4.

![Box-and-whisker plots of resistivities](image)

**Figure 6.** Box-and-whisker plots of the calculated resistivities.

**Table 4.** Obtained resistivities for investigated inks.

| Ink               | Resistivity [µΩ·cm] | Resistivity Related to Bulk Silver |
|-------------------|---------------------|----------------------------------|
| Clariant TPS50    | 3.5 ± 1.6           | 2.2                              |
| UTDots Ag25X      | 14.3 ± 3.6          | 9                                |
| UTDots Ag40X      | 12.3 ± 5.0          | 7.8                              |

The resistivity of bulk silver is 1.59 µΩ·cm [29]. When this was compared with the obtained resistivities for the researched inks, the relative values shown in the third column of Table 4 were obtained.

4. Discussion

An investigation of AZO resistance changes due to thermal curing gives valuable information, which constitutes optimal thermal treatment of overprints. As temperatures higher than 225 °C cause degradation of the AZO layer, it was necessary to find curing conditions that do not damage AZO and make overprints conductive. A curing process of 200 °C for 1 h is a compromise between maintaining the electrical properties of AZO and satisfactory sintering of printed paths. This requirement disqualified other methods...
of thermal curing found in the literature, such as photonic sintering [30], due to their damaging the AZO layer.

Printouts made of TPS50 ink as shown on Figure 3 had a width of 13 µm. Widths of 8 µm were also obtained, but their quality was poorer. In comparison to the results described in the literature, our collecting electrodes can diminish the shadowing level of PV devices through printing thinner lines [15,16]. Printouts are narrower due to optimization of printing parameters and 100 µm printhead utilization, which is one of the smallest available. Eckstein et al. measured conductivity of used materials resulting in a value of $10^7$ S/m order of magnitude, which is adequate to resistivity of 10 µΩ·cm [15]. This value is comparable to the electrical properties of UTDots inks, and three times higher than Clariant resistivity.

Overprints evaluation showed material differences between inks. The printing process resulted in specific intervals of printed-line width for each ink (solvents contribution, functional phase content). UTDots inks are characterized by poorer electrical properties than Clariant Prelect TPS50, but uncertainty is much lower for UTDots inks. This indicates that overprints made with those inks are more homogeneous and the printing process itself is more stable.

Obtained widths of overprints are the same or smaller than those presented in Table 1, which shows a way of improving a photovoltaic’s efficiency and fill factor through modification of collecting electrodes.

5. Conclusions

Commercial inks from Clariant and UTDots were characterized in terms of the best printing parameters, overprints appearance, and material conductivity related to specific thermal curing conditions. The applied process gave high conductivity values (to 0.45 of bulk silver, Clariant). Clariant overprints were found to be much less homogeneous (uncertainty is the same order of magnitude as resistivity value). It was found that a UA results in a more stable printing process, which is crucial to achieve homogeneous and repeatable printouts. The source of the stability was found to lie in the size of the aerosol droplets.

The values of electrical parameters and geometry can be further improved with dedicated inks, which are engineered with consideration of the physical properties of specific substrates. Commercial materials are designed to perform in various applications. This approach to engineering inks leads to good results, but not the best possible. Future research will be focused on testing printed collecting electrodes on working photovoltaic devices to verify whether there is improvement in FF and PCE values.

This study shows that there is a place for AJP technology in decreasing the shadowing level of photovoltaic devices by collecting electrodes. It is a highly competitive technique with other additive manufacturing technologies due to the benefits it offers: high conductivity with overprints of small width. The fact that it can be obtained using curing conditions non-destructive to AZO leads to the conclusion that AJP is applicable in the photovoltaic industry.

Based on the experience gathered during this research, which is reflected in the data in Table 3, UTDots Ag40X gave a superior printing path in the widest spectrum of all considered inks (high process stability), which makes it reliable for industry application.

The biggest challenges with incorporating AJP into the PV industry concern preparing the discussed method for mass production. It demands a highly stable process enabling long-term continuous printing, constant ink supply, and a sound diagnostic system. Another problem that needs resolution is the low printing speed compared with other competing manufacturing methods. Utilization of multiple printheads could address this drawback.
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