EVALUATION OF TEXTURED TCOs FOR a-Si:H/µc-Si:H THIN FILM SOLAR CELLS BY ANGULAR RESOLVED LIGHT SCATTERING MEASUREMENTS

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ABSTRACT

Transparent conductive oxides are often used as transparent front electrodes for thin film solar cells. Besides their conductivity and transparency in the absorption range of the solar cell also the light scattering ability is important for light management. The TCOs are textured, e.g. as grown LPCVD ZnO:B or texture-etched ZnO:Al, and depending on their morphology they are differently adequate for their application as front electrode in solar cells. An evaluation method based on angular resolved light scattering (ARS) is presented in this paper. Measurement results and evaluation of different types of textured TCOs like reactively MF sputtered ZnO:Al (Zn:Al target), RF sputtered ZnO:Al (ZnO:Al on angular resolved light scattering (ARS) is presented in this paper. Measurement results and evaluation of different types of textured TCOs like reactively MF sputtered ZnO:Al (Zn:Al target), RF sputtered ZnO:Al (ZnO:AlO target) and LPCVD ZnO:B are shown. A correlation between ARS and short-circuit density of a-Si:H/µc-Si:H p-i-n solar cells was found for LPCVD ZnO:B and reactively or RF sputtered and etched ZnO:Al until a saturation current was reached.

Keywords: Transparent Conducting Oxides, Zinc Oxide, a-Si/µc-Si, Thin Film Silicon Solar Cell, Light Trapping

1 INTRODUCTION

In the field of thin film silicon photovoltaics an effective light trapping is crucial for a good performance of the solar cell. Therefore textured TCOs are often used as front electrode to obtain a better light incoupling into the cell [1, 2]. The optical pass can be strongly enhanced by light scattering and total internal reflections in the ideal case [3]. The evaluation of textured TCOs concerning light scattering ability is a very important research field. Usually rough and transparent surfaces are characterized by their haze, which is a scalar value and directly connected to the rms-roughness [4,5]. Nevertheless, it was shown, that the haze alone is not sufficient to characterize the quality of a TCO for light trapping in all cases [5].

To include also the lateral structure size into the evaluation, angular resolved light scattering (ARS) is a valuable tool. In the case of rms roughness smaller than 30% of the wavelength it is given by an optical factor multiplied with a surface factor [6].

In the following the cell performance of a-Si:H/µc-Si:H solar cells on different textured TCOs will be examined and put into correlation with large angular scattering of the TCOs.

2 EXPERIMENTAL

2.1 TCO preparation and characterization

Boron doped ZnO was prepared on glass substrates by LPCVD at Malibu. Its morphology was varied by changing film thickness and boron doping. (more details??, eg. precursors used, temperature, pressure)

Reference samples were prepared at Forschungszentrum Jülich (IEK-5) by RF sputtering of ZnO:Al from a ceramic ZnO:AlO target. (P, pressure, T, carrier velocity, target doping, generator, oxygen partial pressure etc. see table 1).

At Fraunhofer IST three reactively sputtered ZnO:Al series were done. Metallic Zn:Al (0.3 wt.%) targets were sputtered with XH frequency excitation at different oxygen partial pressures. Details can be found in [7,8]. Further deposition parameters are summarized in Table 1. The sputtered ZnO:Al was texturized after deposition with an additional etch step in 0.5 wt% HCl. Here 150 nm was taken off the samples. To adjust the transmittance in the near infrared, after etching the samples of series 2 were annealed at 600°C heater temperature in vacuum for 2-2.5 h. This leads to a reduction of free charge carriers and therefore to an increase of transmittance at longer wavelength.

Table 1: Deposition parameters of the used TCOs.

<table>
<thead>
<tr>
<th>PK 750 cathode, MF excitation</th>
<th>90 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>target-substrate distance</td>
<td>90 mm</td>
</tr>
<tr>
<td>generator</td>
<td>MEI, MF, 40 kHz (sine)</td>
</tr>
<tr>
<td>target material</td>
<td>Zn:Al 0.3 wt. % Al</td>
</tr>
<tr>
<td>target size</td>
<td>750x 88 mm³</td>
</tr>
<tr>
<td>Ar-flow</td>
<td>2 x 120 sccm</td>
</tr>
<tr>
<td>mixed gas flow</td>
<td>120 sccm (10% O₂)</td>
</tr>
<tr>
<td>substrate temperature</td>
<td>310-330°C</td>
</tr>
<tr>
<td>chamber pressure</td>
<td>440 mPa</td>
</tr>
<tr>
<td>carrier velocity</td>
<td>3-5.6 mm/s</td>
</tr>
<tr>
<td>discharge power</td>
<td>3.7 – 8.3 kW</td>
</tr>
</tbody>
</table>

http://dx.doi.org/10.4229/26thEUPVSEC2011-3AV.2.43
p (O₂)
series 1 6, 8, 10, 12, 14 mPa
series 2 18, 20, 22, 24, 26, 28 mPa
series 3 6, 8, 10, 16, 20, 22 mPa

All TCOs were optically characterized by transmittance and reflectance measurements with a Varian 5, while the textured samples were measured using an integrating sphere.

To obtain a value expressing the transparency of the TCOs in the absorption range of a microcrystalline bottom cell in an typical a-Si:H/µc-Si:H solar cell on textured ZnO:Al, the transmittance was folded with the external quantum efficiency (compare Figure 1). Therefore the EQE was multiplied with the standard AM1.5G spectrum, the sum was normalized to 1.0 in the wavelength range 300-1100 nm. These values were multiplied with the transmittance and integrated. This gives a first-order approximation for a figure of merit including the transparency of the TCOs in the relevant wavelength range. Of course transmittance at air and in a solar cell with adjacent silicon is different.

The textured TCOs were examined with angular resolved light scattering (ARS) using an ARTA accessory for a Perkin Elmer Lambda 950 supplied by OMT Solutions [9]. The samples were illuminated from the coated side at different wavelengths and a photomultiplier collected the scattered light intensity at angles in the plane of incidence [10]. Step width was chosen to be 2°, the detector opening was 3x17 mm². The baseline was collected with the same opening, cutting the incident beam spot to one third, which has the effect of an attenuator increasing the measured intensity by a factor of three. Assuming the sample is scattering isotropically in all directions, the signal was weighted according to the scattering angle θ. The weighting factor is proportional to sin(θ).

2.2 Solar cell preparation and characterization
a-Si:H/µc-Si:H solar cells were deposited in pin configuration on glass substrates. The cell depositions were done by our project partners Forschungszentrum Jülich (IEK-5) (series 1 and 2) and Malibu (LPCVD ZnO:B) by PECVD. Absorber thicknesses and substrates are given in table 2.

Table II: Solar cell series.

<table>
<thead>
<tr>
<th>series</th>
<th>absorber thickness</th>
<th>substrate</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-Si:H</td>
<td>µc-Si:H</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>300 nm 1.3 µm</td>
<td>SGG Diamant + SiO,N,</td>
</tr>
<tr>
<td>2</td>
<td>350 nm 1 µm</td>
<td>Corning 1737</td>
</tr>
<tr>
<td>3</td>
<td>300 nm 1 µm</td>
<td>Optiwithe + SiO,N,</td>
</tr>
<tr>
<td></td>
<td>300 nm 1.6 µm</td>
<td>Optiwithe + SiO,N,</td>
</tr>
<tr>
<td>LPCVD</td>
<td>256 nm 1.6 mm</td>
<td>White glass</td>
</tr>
</tbody>
</table>

Current-voltage-curve (IV) measurements and external quantum efficiency (E QE) measurements of the cells were done under standard conditions at the involved laboratories.

3 RESULTS AND DISCUSSION

As a first step as grown RF sputtered ZnO:Al and textured ZnO:Al were compared in their EQE. As can be seen in Figure 1 the EQE could be drastically enhanced especially in the bottom cell. Here light trapping is very important due to the low extinction coefficient of µc-Si. By texturing the TCO, the short circuit current density \( J_{sc} \) could be increased from 10.7 to 11 mA/cm² for the a-Si:H top cell and from 7.6 to 11.5 mA/cm² for the µc-Si:H bottom cell. This is an increase of over 50% in \( J_{sc} \).

When etching ZnO:Al samples the etching occurs randomly. Craters are forming as can be seen in Figure 2 and Figure 5. Depending on dynamical deposition and/or dynamical etching, anisotropical etch morphologies can occur, which are demonstrated in the SEM pictures in Figure 2. The horizontal anisotropy is only observed in B orientation (see Figure 2, left), rotated clockwise by 90° the craters appear symmetrical again (orientation A). This can be confirmed by ARS measurements (Figure 3). For sample orientation B the ARS signal is symmetrical. If the sample is rotated counterclock- (A) and clockwise (C) by 90° the ARS signal reveals the anisotropy.

![Figure 1: EQE of a-Si:H/µc-Si:H solar cells (350 nm a-Si, 1µm µc-Si) with etched and unetched RF sputtered ZnO:Al front contact.](image1)

![Figure 2: SEM pictures of etched ZnO:Al with anisotropic etch morphology. Left: top view, 5,000X. The sample orientation for ARS measurements is shown. Right: Sample turned by 90° clockwise: 10,000X, 60° tilt.](image2)
In the case of anisotropy no sine-weighting as described before should be done. For the reactively sputtered and etched ZnO:Al and also for LPCVD ZnO:B no anisotropy in scattering was observed, therefore in the following only sine-weighted data will be shown.

As can be seen in Figure 1, light trapping is most relevant for the bottom cell, whose absorption maximum is around 750 nm. Therefore the light scattering was measured around that wavelength. 700 nm was chosen, because at 750 nm the signal is very noisy for these low intensities. The ARS signal of reactively sputtered and etched ZnO:Al (series 1) is shown in Figure 4. With increasing oxygen partial pressure the samples tend to exhibit rougher and sharper etch morphologies (compare figure 5 and [7]), which lead to increased large angle scattering (>46°). The large etch craters of RF sputtered ZnO:Al, which is used as reference here, pronounce scattering also into small angles around 12°.

The ARS signal was integrated between the angles 46-80°. Then $J_{sc, \text{bottom}}$ determined from the µc-Si bottom EQE of the corresponding a-Si/µc-Si solar cell over large angular scattering (46-80°).

Two factors influence $J_{sc}$: Mainly it will be the light scattering leading to an efficient light trapping. The second factor is the transmittance of the TCO itself. The less light passes through the TCO, the lower $J_{sc}$ will be. In Figure 5 the transmittance of the ZnO:Al of series 1 is shown. At 1150 nm a big difference can be observed in transmittance between the different samples. Therefore it is necessary to normalize $J_{sc, \text{bottom}}$ to the transmittance in that wavelength range to reveal the influence of morphology only. $T_{\text{EQE, bottom}}$ was calculated as described in the experimental section.

Firstly, for all deposited TCO samples the $J_{sc, \text{bottom}}$ of the corresponding solar cells was plotted against the integrated ARS signal (46-80°) at 700 nm (Figure 6). A linear correlation could be seen for series 1, 3 and the LPCVD ZnO:B, while the latter showed a lower slope. For series 2 no increase of $J_{sc, \text{bottom}}$ with large angle scattering could be observed any more. A maximum $J_{sc, \text{bottom}}$ around 11.4 mA/cm² was reached here. Maybe the silicon growths introduces more defects acting as recombination centers when a certain substrate roughness is exceeded. At this point light scattering cannot improve $J_{sc, \text{bottom}}$ further on.
The thicker the µc-Si absorber, the higher is $J_{sc, bottom}$ even for lower ARS $>46^\circ$. The slope seems simply to be shifted to higher $J_{sc}$. For series 3 no EQE data was available yet, and IV-data was used, which is only reliable for bottom-limited tandem solar cells. Therefore the data for the thick 1.6 µm µc-Si absorber is less reliable.

Figure 6: $J_{sc}$ in dependence of large angle scattering of the TCO for different a-Si/µc-Si solar cell series either deposited onto reactively sputtered and etched ZnO:Al or LPCVD ZnO:B. $J_{sc}$ was determined by EQE of the µc-Si bottom cell if available or by IV-measurements.

To distinguish between morphology and transmittance effects, the data of Figure 6 was normalized to the transmittance of the TCOs, respectively (Figure 7). It can be seen that for thin µc-Si absorbers in series 1 and 3 and also for the LPCVD ZnO:B there is still a linear correlation visible. This is a sign that morphology is the main reason for an increase in $J_{sc, bottom}$ here. The opposite occurred for the thick absorber in series 3. Here no dependence on scattering can be observed anymore.

Figure 7: Data of Fig.6 normalized according to the TCO transmittance in the range of the µc-Si bottom cell absorption. Transmittance of the unetched samples was therefore folded with the EQE. This was done to separate the effect of different transmittance and light scattering properties of the TCO morphologies.

Figure 8 shows the efficiency of the fabricated a-Si:H/µc-Si:H solar cells. For series 1 and 3 it increases with increasing large angle scattering, which can be attributed to the increase of $J_{sc, bottom}$. Series 3 shows a decline in efficiency with increasing light scattering although $J_{sc, bottom}$ is constant.

Figure 8: Efficiency of the solar cell series. With increasing large angle scattering efficiency increases but the falls again.

An explanation is supported by Figure 9, which shows the open circuit voltages of the solar cells. Here series 1 and 2 exhibit a decreasing $V_{oc}$ with increasing light scattering (which itself is correlated to rougher and sharper morphologies in the case of reactively sputtered ZnO:Al). Therefore the efficiency of series 2 declines because of falling $V_{oc}$, while for the other series (1-3) $J_{sc, bottom}$ is much more increasing than $V_{oc}$ is falling. Cells with thicker µc-Si:H absorbers do not show a decline in $V_{oc}$.

Figure 9: Open circuit voltage of the solar cell series. It is falling for higher large angle scattering (implying steeper morphologies) if the µc-Si absorber is < 1.3 µm.

Because cell depositions and measurements were done at different laboratories, there might be more scattering in the data than expected.

4 CONCLUSION

It was shown, that the use of textured TCOs increases $J_{sc, bottom}$ of the µc-Si bottom cell in a-Si:H/µc-Si:H by about 50%. Angular resolved light scattering is a suitable tool to reveal anisotropic etch morphologies. A correlation between large angle scattering and short-
circuit density of a-Si:H/μc-Si:H solar cells was found for LPCVD ZnO:B and reactively sputtered and etched ZnO:Al. Therefore it is possible to predict the short circuit current of a a-Si:H/μc-Si:H solar cell for a given absorber thickness and type of TCO morphology just from ARS measurements. This works only until a maximum J_{sc} is reached. Here other loss mechanisms, e.g. recombination in defects, seem to become the dominant limitation process.

A decrease in V_{oc} was observed for thin μc-Si absorbers if the TCO morphology shows sharper structures. This can be overcome with smoother structures or thicker absorbers.

The TCO transmittance in the NIR limits J_{sc}, bottom to a certain level. For reactively sputtered ZnO:Al theoretically 1-2 mA/cm² could be won, for LPCVD ZnO:B even 3 mA/cm², if transmittance could be enhanced ideally over the whole wavelength range.

5 REFERENCES


7 ACKNOWLEDGEMENT

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