Improved surface passivation and reduced parasitic absorption in PEDOT:PSS/c-Si heterojunction solar cells through the admixture of sorbitol

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We examine the impact of sorbitol admixture to the hole-conduction polymer PEDOT:PSS [poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)] on the characteristics of PEDOT:PSS/crystalline silicon heterojunction solar cells. We fabricate solar cells where the PEDOT:PSS layer is deposited as a hole-collecting contact at the cell rear, whereas the electron-collecting front is conventionally processed by means of phosphorus diffusion. Surprisingly, we observe that the admixture of the infrared-transparent sorbitol not only improves the short-circuit density of the solar cells due to the reduction of the infrared parasitic absorption, but also improves the passivation quality of PEDOT:PSS on silicon and hence the open-circuit voltage of the solar cells. The series resistance is not influenced by the admixture of sorbitol up to 4.0 wt.% sorbitol admixture in the PEDOT:PSS dispersion, but shows a pronounced increase for larger sorbitol contents. The optimal sorbitol content concerning efficiency is hence 4.0 wt.%, leading to an energy conversion efficiency of 20.4% at one sun, which is more than 1% absolute higher compared to the efficiency of the reference cells without sorbitol.

Heterojunction solar cells combining the hole-conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) [PEDOT:PSS] and c-Si have demonstrated their high-efficiency potential in previous studies1–9. In the first fabricated PEDOT:PSS/c-Si solar cells, the organic layer was deposited to the front, leading to relatively low short-circuit current densities \(J_{sc}\) due to the parasitic absorption losses within the PEDOT:PSS. Moving the organic layer to the rear in the so-called ‘BackPEDOT’ cell concept10 resulted in significantly improved \(J_{sc}\) and led for the first time to efficiencies exceeding 20%11,12. Still it turned out that even for the BackPEDOT cell, \(J_{sc}\) is limited by parasitic free-carrier absorption within the PEDOT:PSS layer of infrared photons reaching the cell rear10. In the present study, we examine a promising and easy-to-implement approach to reduce the parasitic absorption in BackPEDOT cells by increasing the transparency of the PEDOT:PSS layer by adding sorbitol to the precursor dispersion. In a previous study13, we showed that sorbitol significantly increases the transparency of the PEDOT:PSS layer in the infrared and thus sorbitol can be used for effectively reducing the parasitic absorption in the PEDOT:PSS layer. Sorbitol is already established as a conductivity additive in PEDOT:PSS14,15. In this work, we fabricate solar cells to examine the impact of the sorbitol admixture on device level. In addition, we also examine the impact on surface passivation on lifetime samples. Our experimental results clearly show that the sorbitol admixture does not only positively affect the \(J_{sc}\) of the cells, but due to an improved surface passivation, shows also a positive impact on the open-circuit voltage \(V_{oc}\). Hence, the addition of sorbitol to the PEDOT:PSS dispersion has multiple advantages for PEDOT:PSS/c-Si heterojunction cells.
Experimental Details

Lifetime sample preparation. In order to assess the passivation quality of the PEDOT:PSS/c-Si interface, contactless lifetime samples are fabricated on 300 μm thick (100)-oriented p-type float-zone silicon (FZ-Si) wafers with a resistivity of 200 Ωcm. After RCA cleaning, one wafer surface is passivated by a 100 nm thick plasma-enhanced-chemical-vapor-deposited (PECVD) SiNx layer (Plasmalab 80 Plus, Oxford) with a refractive index of 2.4 at a deposition temperature of 400 °C. After SiNx deposition, the samples are dipped in 1% hydrofluoric acid (HF) for 60 seconds. The PEDOT:PSS dispersion (Clevios special grade, Clevios Heraeus GmbH), which has a solid content of 2.2 wt. % of PEDOT:PSS in water, was mixed with 1.0 to 18.0 wt.% sorbitol and stirred overnight at room temperature on a magnetic stirrer. Immediately after the HF dip, the PEDOT:PSS dispersion is deposited by spin coating at 500 revolutions per minute (rpm) for 10 seconds and subsequently 1500 rpm for 30 seconds. Subsequently, the PEDOT:PSS layer is annealed in ambient environment at a temperature of 130 °C for 10 min. Injection-dependent measurements of the effective carrier lifetime \(\tau_{\text{eff}}(\Delta n)\) are performed using a Sinton Lifetime Tester (WCT-120, Sinton Instruments). The recombination current density parameter (also sometimes denoted ‘saturation current density’) \(J_0\) of each measured sample is extracted from the slope of the inverse lifetime \(1/\tau_{\text{eff}}\) vs. the excess carrier density \(\Delta n\) curve.

Solar cell fabrication. Figure 1 shows the cross-section of a BackPEDOT solar cell. For the solar cell fabrication we use 300 μm thick 6″ (100)-oriented p-type boron-doped FZ-Si wafers with resistivities of 0.5 and 1.3 Ωcm, respectively. After RCA cleaning and protecting both surfaces by a 200 nm thick dielectric SiO2 layer, 2 × 2 cm2 diffusion windows are opened by laser ablation on one wafer surface. After one more RCA cleaning sequence, the silicon surface within the ablated window is random-pyramid-textured using a KOH/isopropanol solution. Subsequently, an n+ emitter with a sheet resistance in the range 92–110 Ω/sq is formed by phosphorus in-diffusion ([P]) are performed using a Sinton Lifetime Tester (WCT-120, Sinton Instruments). The PEDOT:PSS precursor dispersion is deposited by spin coating at 500 rpm for 10 seconds and subsequently 1500 rpm for 30 seconds. Subsequently, the PEDOT:PSS layer is annealed in ambient environment at a temperature of 130 °C for 10 min. Injection-dependent measurements of the effective carrier lifetime \(\tau_{\text{eff}}(\Delta n)\) are performed using a Sinton Lifetime Tester (WCT-120, Sinton Instruments). The recombination current density parameter (also sometimes denoted ‘saturation current density’) \(J_0\) of each measured sample is extracted from the slope of the inverse lifetime \(1/\tau_{\text{eff}}\) vs. the excess carrier density \(\Delta n\) curve.

Results and Discussion

Surface passivation. Figure 2 shows a box plot of the measured recombination current density parameter \(J_0\) of the PEDOT:PSS/c-Si interface as a function of the sorbitol content added to the PEDOT:PSS precursor dispersion.

Table 1 shows the PEDOT:PSS layer thicknesses of the lifetime samples shown in Fig. 2. The corresponding total sheet resistances are also shown in Table 1. The reduction of the sheet resistance by adding more than 1.0 wt.% sorbitol agrees well with the behaviour reported in the literature that sorbitol serves as a conductivity-increasing additive14,15. The PEDOT:PSS layer thicknesses were determined using a profilometer (Dektak 150, Vecco) and the corresponding sheet resistances were measured using a Sinton lifetime tester (WCT-120, Sinton Instruments). \(J_0\) was extracted from the measured overall \(J_0\) values by subtraction of the \(J_{\text{SSN}}\) value of the SiNx-passivated wafer surface. \(J_{\text{SSN}}\) was determined on both-sides-SiNx-passivated wafers with a median value of 16 fA/cm2. For silicon surfaces passivated with PEDOT:PSS without admixture of sorbitol, we measure a median \(J_{\text{PEDOT}}\) value of 86 fA/cm2. We observe that with increasing sorbitol content of the PEDOT:PSS precursor dispersion the measured \(J_{\text{PEDOT}}\) decreases. At a sorbitol concentration of 4.0 wt.% in the precursor dispersion, we measure a median \(J_{\text{PEDOT}}\) value of 64 fA/cm2. At a sorbitol concentration of 7.7 wt.%, a median \(J_{\text{PEDOT}}\) of only 38 fA/cm2 is achieved, which means that the recombination at the PEDOT:PSS/c-Si interface is more than halved by adding 7.7 wt.% of sorbitol to the precursor solution. At even higher sorbitol concentrations of 11 wt.% and 14 wt.% the \(J_{\text{PEDOT}}\) value rises to a median of 55 fA/cm2 and 58
A sorbitol concentration of 18 wt.% provides a $J_0$ median of 49 fA/cm$^2$. According to our results a saturation in $J_0$ at high sorbitol concentrations can be observed. Nardes et al. have shown that the addition of sorbitol reduces the work function of PEDOT:PSS for sorbitol concentration of 5 wt.%, and for higher sorbitol concentrations a saturation in the work function occurs. The positive impact of sorbitol on the electronic passivation properties of PEDOT:PSS on c-Si might be due to a change in the electronic band structure of the organic solution. It is well known from the literature that the addition of sorbitol to the PEDOT:PSS precursor dispersion reduces the work function of the PEDOT:PSS with increasing sorbitol content. Our results here clearly prove that adding sorbitol to the PEDOT:PSS precursor solution significantly improves the c-Si surface passivation quality.

Solar cells. A total of 42 PEDOT:PSS/c-Si solar cells with an area of $2 \times 2$ cm$^2$ were fabricated. The precursor dispersion of the spin-coated PEDOT:PSS layer on the solar cell rear was mixed with different contents of sorbitol and stirred overnight on a magnetic stirrer. We examine the impact of the sorbitol admixture on the most relevant solar cell parameters that are the open-circuit voltage $V_{oc}$, the short-circuit current density $J_{sc}$, the fill factor $FF$ and the series resistance $R_s$ as well as the energy conversion efficiency $\eta$. The $J_{sc}-V_{oc}$ characteristics as well as the illuminated current-voltage ($J-V$) curves are measured under standard testing conditions at 1 sun and 25°C using a LOANA measurement system (pv-tools, Hamelin, Germany). The series resistance $R_s$ is determined from the measured fill factor $FF$, pseudo-fill factor $pFF$ as well as the measured $J_{sc}$ and $V_{oc}$ values using the equation:

![Figure 2. Box plot of the recombination current density parameter $J_{0,PEDOT}$ of the PEDOT:PSS/c-Si interface as a function of the sorbitol content of the PEDOT:PSS precursor dispersion. PEDOT:PSS was applied to 9 test samples for each sorbitol concentration. The $J_{0,SiN}$ value of 16 fA/cm$^2$ of the SiN$_x$-passivated silicon surface was subtracted from the measured $J_{0}$ value to account only for recombination at the PEDOT:PSS/c-Si interface.]

| Sorbitol content in the PEDOT:PSS precursor dispersion [wt.%] | PEDOT:PSS layer thickness [nm] | Total sheet resistance [Ω/ ] |
|-------------------------------------------------------------|--------------------------------|-----------------------------|
|                 | PEDOT:PSS applied at 1500 rpm | PEDOT:PSS applied at 12000 rpm | PEDOT:PSS applied at 1500 rpm | PEDOT:PSS applied at 12000 rpm |
| 0.0             | 1674 ± 343                    | 181 ± 60                     | 2269                        | 2518                          |
| 1.0             | 978 ± 123                     | 140 ± 36                     | 1851                        | 2056                          |
| 2.0             | 1070 ± 125                    | 146 ± 32                     | 124                         | 2083                          |
| 4.0             | 1623 ± 240                    | 162 ± 41                     | 67.3                        | 435                           |
| 5.7             | 2137 ± 237                    | 163 ± 39                     | 61.5                        | 304                           |
| 7.7             | 2087 ± 322                    | 184 ± 55                     | 60.2                        | 274                           |
| 11.0            | 1033 ± 47                     | —                            | 99.2                        | —                             |
| 14.0            | 1766 ± 52                     | —                            | 76.6                        | —                             |
| 18.0            | 1936 ± 81                     | —                            | 79.6                        | —                             |
As the open-circuit voltage $V_{oc}$ is very sensitive to recombination losses, the positive impact of the sorbitol admixture observed on the lifetime test samples shown in Fig. 2, should translate to increased $V_{oc}$ values on the solar cells with sorbitol admixture. Figure 3(a) shows the measured $V_{oc}$ values of each processed BackPEDOT solar cell as a function of the sorbitol concentration in the PEDOT:PSS precursor dispersion. The median parameters of the solar cells with 0.5 Ωcm base resistivity are shown in Table 2 and the parameters of the 1.3 Ωcm cells in Table 3.

As can be seen from Fig. 3(a) and Tables 2 and 3, $V_{oc}$ increases from a median value of 655 mV to 659 mV for the 0.5-Ωcm p-Si base material and from 657 mV to 660 mV for the 1.3-Ωcm base material by adding 4.0 wt.% of sorbitol to the PEDOT:PSS precursor dispersion. At a sorbitol concentration of 5.7 wt.%, the median $V_{oc}$ values decrease to 650 mV for the 0.5-Ωcm cells and to 643 mV for the 1.3-Ωcm cells. While the improvement in $V_{oc}$ by the admixture of up to 4.0 wt.% of sorbitol is in good agreement with our lifetime study shown in Fig. 2, the reduced $V_{oc}$ at 5.7 wt.% admixture of sorbitol is unexpected and somewhat peculiar, but seems to be reproducible, as it was observed in all eight cell batches processed within this work. Increasing the sorbitol content further to 7.7 wt.% increases the median $V_{oc}$ approximately back to the state without sorbitol admixture. Nevertheless, the maximum $V_{oc}$ value of a single cell of 661 mV was achieved on a 0.5-Ωcm cell with 7.7 wt.% sorbitol admixture. From our experimental results obtained on 42 processed BackPEDOT cells we hence conclude that the admixture of sorbitol can have a clear positive impact on the passivation quality of the PEDPOT:PSS/c-Si junction, although not too much of sorbitol should be added. Figure 3(b) compiles the measured $J_{sc}$ values of all processed PEDOT:PSS/c-Si solar cells as a function of the sorbitol concentration (median values listed in Tables 2 and 3). The solar cells with 1.3-Ωcm base material provide higher $J_{sc}$ values compared to the solar cells fabricated on 0.5-Ωcm base material, which can be attributed to the much higher bulk lifetime in the silicon material with lower doping concentration. Lifetime measurements performed on the 0.5-Ωcm p-type FZ-Si material verified that the
The bulk lifetime of this material is 0.77 ms, whereas that of the 1.3-Ωcm material is 1.56 ms at an excess carrier density $\Delta n$ of $3 \times 10^{15}$ cm$^{-3}$. For both base materials, the addition of sorbitol to the PEDOT:PSS is positively affecting the $J_{sc}$ value. For the 0.5-Ωcm material, the median $J_{sc}$ increases by 0.7 mA/cm$^2$ by adding a sorbitol concentration of 4.0 wt.% to the PEDOT:PSS dispersion. At a higher sorbitol content of 7.7 wt.% the median $J_{sc}$ increases even more by 0.9 mA/cm$^2$ compared to the reference case without sorbitol. For the BackPEDOT cells fabricated on the 1.3-Ωcm $p$-Si base material, the median $J_{sc}$ increases by 0.8 mA/cm$^2$ for a sorbitol concentration of 7.7 wt.% compared to the reference without sorbitol. In this study, the highest measured $J_{sc}$ on a PEDOT:PSS/c-Si solar cell with a base resistivity of 1.3 Ωcm is 39.6 mA/cm$^2$ for a sorbitol content of 7.7 wt.% of the PEDOT:PSS precursor dispersion. From these results it becomes obvious that the addition of sorbitol to the PEDOT:PSS dispersion effectively reduces the parasitic absorption losses in PEDOT:PSS/c-Si heterojunction cells.

Figure 4 shows the measured change in the internal quantum efficiencies $\Delta\text{IQE}$ (blue circles and red triangles) and change in reflectance (blue squares and red diamonds) of exemplary BackPEDOT solar cells by adding 4.0 wt.% (red symbols) and 7.7 wt.% (blue symbols) sorbitol compared to a BackPEDOT solar cell without sorbitol. The solar cells have a $p$-type $c$-Si base with a resistivity of 0.5 Ωcm and the PEDOT:PSS was spin-coated at 500 rpm for 10 seconds and subsequently at 12000 rpm for 30 seconds.

![Figure 4](https://example.com/figure4.png)

Figure 4. Measured change in the internal quantum efficiency ($\Delta\text{IQE}$) (blue circles and red triangles) and change in reflectance (blue squares and red diamonds) of exemplary BackPEDOT solar cells by adding 4.0 wt.% (red symbols) and 7.7 wt.% (blue symbols) sorbitol compared to a BackPEDOT solar cell without sorbitol. The solar cells have a $p$-type $c$-Si base with a resistivity of 0.5 Ωcm and the PEDOT:PSS was spin-coated at 500 rpm for 10 seconds and subsequently at 12000 rpm for 30 seconds.

| Sorbitol content in the PEDOT:PSS precursor dispersion [wt.%] | $V_{oc}$ [mV] | $J_{sc}$ [mA/cm$^2$] | FF [%] | $\eta$ [%] | $R_s$ [Ωcm$^2$] |
|---|---|---|---|---|---|
| Median Max. Median Max. Median Max. Median Max. Median Max. Median Min. |
| 0.0 (2 cells) | 657 | 657 | 38.6 | 38.8 | 75.1 | 75.6 | 19.0 | 19.3 | 1.55 | 1.48 |
| 1.0 (1 cells) | 657 | 657 | 39.0 | 39.0 | 78.9 | 78.9 | 20.2 | 20.2 | 0.86 | 0.86 |
| 2.0 (2 cells) | 659 | 660 | 39.0 | 39.1 | 75.4 | 75.7 | 19.4 | 19.4 | 1.47 | 1.42 |
| 4.0 (1 cells) | 660 | 660 | 39.0 | 39.0 | 78.6 | 78.6 | 20.3 | 20.3 | 0.89 | 0.89 |
| 5.7 (3 cells) | 643 | 655 | 38.5 | 39.0 | 72.6 | 76.5 | 17.9 | 19.5 | 1.98 | 1.27 |
| 7.7 (3 cells) | 656 | 659 | 39.5 | 39.5 | 71.1 | 74.0 | 18.5 | 19.3 | 2.51 | 1.92 |

Table 3. Median and maximum values of the cell parameters for BackPEDOT solar cells with a base resistivity of 1.3 Ωcm.

![Table 3](https://example.com/table3.png)

Table 3. Median and maximum values of the cell parameters for BackPEDOT solar cells with a base resistivity of 1.3 Ωcm.
the electron beam evaporation in combination with a presumably remaining sorbitol content in the PEDOT:PSS layer. Optical microscope images of the metallized cell rear for the different sorbitol concentrations are shown in Supplementary Fig. S1. The amount of sorbitol remaining in the PEDOT:PSS layer is probably higher for sorbitol contents greater than 4 wt. % than for lower sorbitol concentrations. Therefore, we observe an increase in $R_s$ only at high sorbitol concentrations. The best $R_s$ value realized on a solar cell processed in this study is 0.6 Ωcm$^2$ for a sorbitol concentration of 4.0 wt. %, which led to an efficiency of 20.2%. The achieved efficiencies of all fabricated BackPEDOT solar cells as a function of the sorbitol concentration are shown in Fig. 5(b). For PEDOT:PSS/c-Si solar cells manufactured with the PEDOT:PSS dispersion without addition of sorbitol, efficiencies range from 18.4 to 19.7%. By adding a sorbitol content to the precursor dispersion of up to 4.0 wt. %, higher cell efficiencies of up to 20.4% are achieved. If a sorbitol content of more than 4.0 wt. % is added to the precursor dispersion, the cell efficiencies decrease and range from 17.9% to 19.2% at a sorbitol content of 7.7 wt. %. The decrease in solar cell efficiencies with higher sorbitol content is attributed to the increasing $R_s$ value. The best efficiency measured in this study on a PEDOT:PSS/c-Si solar cell is 20.4% with a sorbitol content in the precursor dispersion of 4.0 wt. %.

The illuminated $J-V$ curve and the $J_{sc}-V_{oc}$ curve of the best solar cell of this study with the corresponding solar cell parameters is shown in Fig. 6. The best BackPEDOT solar cell fabricated in this study, shown in Fig. 6, provides a $V_{oc}$ of 660 mV and a $J_{sc}$ value of 38.5 mA/cm$^2$. A good fill factor of 80.0% is achieved due to a low $R_s$ value of 0.73 Ωcm$^2$. This combination of cell parameters leads to the highest efficiency achieved in this study of 20.4% for a PEDOT:PSS/c-Si solar cell with admixture of sorbitol to the precursor dispersion. The $J_{sc}$ value of 38.5 mA/cm$^2$ achieved on this cell seems relatively low and higher $J_{sc}$ values (up to 39.6 mA/cm$^2$) were achieved on the 1.3 Ωcm $p$-Si base material, featuring a much higher bulk lifetime compared to the 0.5 Ωcm bulk material. Hence, there seems to be a strong contribution of the bulk recombination in the record cell of our current study and the use of better bulk material should allow to further improve the efficiency.

Conclusions
In this study, we have fabricated 42 solar cells with phosphorus-diffused front and PEDOT:PSS/c-Si heterojunction at the rear (‘BackPEDOT’ cells). We have examined the impact of adding sorbitol to the PEDOT:PSS precursor dispersion on the solar cell parameters. The recombination current density parameter $J_{0,\text{PEDOT}}$ of the PEDOT:PSS/c-Si interface showed a median $J_{0,\text{PEDOT}}$ value of 86 fA/cm$^2$ for silicon surfaces passivated with PEDOT:PSS without admixture of sorbitol. At a sorbitol concentration of 7.7 wt. %, the median $J_{0,\text{PEDOT}}$ value was lowered to only 38 fA/cm$^2$ which means that the recombination at the PEDOT:PSS/c-Si solar cell is effectively suppressed by the addition of sorbitol. We also observed that the admixture of sorbitol improved the $J_{sc}$ values due to the reduced parasitic absorption within the PEDOT:PSS layer, by 0.9 mA/cm$^2$ at a sorbitol concentration of 7.7 wt. % on solar cells with a $p$-Si base resistivity of 0.5 Ωcm. The highest achieved $J_{sc}$ value was 39.6 mA/cm$^2$ on a solar cell with a $p$-Si base resistivity of 1.3 Ωcm and a sorbitol concentration of 7.7 wt. %. Due to the improvement in the surface passivation quality due to the sorbitol admixture, we observed an increase in $V_{oc}$ of 4.0 mV on solar cells with a base resistivity of 0.5 Ωcm. An increase of 3.0 mV was observed for solar cells with 1.3 Ωcm base resistivity for admixture of sorbitol contents of 4.0 wt. % to the PEDOT:PSS dispersion. The maximum $V_{oc}$ value of a single cell of 661 mV was achieved on a 0.5-Ωcm cell with 7.7 wt. % sorbitol admixture. The series resistance $R_s$ did not change by up to 4.0 wt. % of sorbitol admixture, but showed a pronounced increase at higher sorbitol concentrations. The optimal sorbitol content concerning efficiency is hence 4.0 wt. %, leading to a median efficiency.
of 20.2% (5 cells), which is 1.1% absolute higher compared to the median efficiency of the reference cells without sorbitol. The highest efficiency achieved in this study was 20.4% for a sorbitol content of 4.0 wt.%. Our experimental results hence suggest that the admixture of sorbitol increases the efficiency potential PEDOT:PSS/c-Si heterojunction solar cells.

Data Availability
All data generated or analyzed during this study are included in this published article.

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Author Contributions
M.-U.H. collected and analyzed the measurement data and wrote the manuscript under the guidance of J.S. D.Z. and R.G. have established and optimized the process flow. R.S.-S. and W.L. produced and provided the PEDOT:PSS dispersion.

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