Efficiency Enhancement in Organic Photovoltaic Cells: Consequences of Optimizing Series Resistance

By Jonathan D. Servaites, Sina Yeganeh, Tobin J. Marks,* and Mark A. Ratner*

Here, means to enhance power conversion efficiency (PCE or $\eta$) in bulk-heterojunction (BHJ) organic photovoltaic (OPV) cells by optimizing the series resistance ($R_s$)—also known as the cell internal resistance—are studied. It is shown that current state-of-the-art BHJ OPVs are approaching the limit for which efficiency can be improved via $R_s$ reduction alone. This evaluation addresses OPVs based on a poly(3-hexylthiophene):6,6-phenyl C$_{61}$-butyric acid methyl ester (P3HT:PCBM) active layer, as well as future high-efficiency OPVs ($\eta > 10\%$). A diode-based modeling approach is used to assess changes in $R_s$. Given that typical published P3HT:PCBM test cells have relatively small areas (~0.1 cm$^2$), the analysis is extended to consider efficiency losses for larger area cells and shows that the transparent anode conductivity is then the dominant materials parameter affecting $R_s$ efficiency losses. A model is developed that uses cell sizes and anode conductivities to predict current–voltage response as a function of resistive losses. The results show that the losses due to $R_s$ remain minimal until relatively large cell areas (>0.1 cm$^2$) are employed. Finally, $R_s$ effects on a projected high-efficiency OPV scenario are assessed, based on the goal of cell efficiencies >10%. Here, $R_s$ optimization effects remain modest; however, there are now more pronounced losses due to cell size, and it is shown how these losses can be mitigated by using higher conductivity anodes.

1. Introduction

Organic photovoltaic (OPV) cells\cite{1–5} offer the potential to change our energy landscape due to low production costs, mechanical flexibility, and the versatility of organic materials design.\cite{6,7} However, while there have been significant gains in OPV cell power conversion efficiencies, they have not reached the level of conventional inorganic photovoltaics.\cite{8,9} State-of-the-art bulk heterojunction (BHJ)\cite{10,11,12} OPVs currently achieve cell efficiencies up to ~6% under the standard solar spectrum, AM1.5G.\cite{4,6,11–15} However, significant improvements are still needed, with efficiencies of 10% or greater generally regarded as necessary for widespread commercial viability. Various useful approaches to BHJ OPV designs have been employed, such as using novel organic materials\cite{4,11,12,17–22} or organic–inorganic hybrids\cite{23,24} as active layer materials, incorporating designed interfacial layers at either electrodes\cite{14,15,25} and employing tandem cells.\cite{13} While these approaches offer significant advantages, loss mechanisms in OPVs still are not completely articulated. Establishing an improved understanding of these losses should provide research focus in increasing power conversion efficiencies to 10% and greater.

One of the key solar cell parameters affecting the cell power conversion efficiency (PCE or $\eta$) is the series resistance, or internal resistance, $R_s$. The series resistance represents the total resistance of the cell and is a composite of: 1) the active and interfacial layer resistances, 2) electrode resistances, and 3) the various contact and interconnect resistances.\cite{26,27} It is known that $R_s$ can have a pronounced effect on the solar cell fill factor ($\beta_{FF}$), which is defined as: \cite{26–29}

$$\beta_{FF} = \frac{J_{max} V_{max}}{J_{sc} V_{oc}} \quad (1)$$

where $J_{max}$ and $V_{max}$ are the current density and voltage, respectively, at the maximum power output of the solar cell, $J_{sc}$ is the cell short circuit current density, and $V_{oc}$ is the cell open circuit voltage. The fill factor is one of three primary factors in determining $\eta$: \cite{26–28}

$$\eta = \frac{\beta_{FF} J_{sc} V_{oc}}{P_{solar}} \quad (2)$$

where $P_{solar}$ is the incident solar radiation. $R_s$ is often the dominant parameter determining the fill factor (by altering the slope of the current density–voltage ($J$–$V$) curve near the $V_{oc}$)\cite{28} and is therefore of key importance for high-efficiency solar cells.

Organic semiconductors used in solar cells have mobilities typically several orders of magnitude lower than their inorganic
counterparts (e.g., silicon, gallium arsenide). Therefore, active layer mobility enhancement and \( R_s \) reduction have been major OPV research focus areas in recent years. Previous studies have demonstrated significant reductions in OPV \( R_s \) values using new materials or fabrication techniques and concluded that the observed efficiency increases are largely due to \( R_s \) reduction. Thus, Li et al. demonstrated significant improvements in \( R_s \) and \( \eta \) for BHJ OPVs by optimizing film growth rates, showing that more slowly grown, phase-separated films benefit from improved molecular self-organization and, consequently, improved hole mobilities. The benefits of self-organization from slow film growth rates have been characterized, as well as the morphological effects of different solvents and annealing rates. However, despite the significant improvements in BHJ OPV mobilities, Li et al. reported maximum time-of-flight mobilities on the order of only \( 10^{-4} \) cm\(^2\) V\(^{-1}\) s\(^{-1}\)—several orders of magnitude less than those of typical inorganic solar cell and organic transistor materials. Furthermore, the resistance of the active layer is proportional to the inverse of the mobility. Therefore, the currently large differences in mobilities between organic and inorganic solar cell active layer materials raise the question of whether and to what degree OPV cell efficiencies suffer from large \( R_s \) values.

While previous authors have concluded that increases in active layer mobilities are correlated with lower \( R_s \) values and, ultimately, greater cell efficiencies, there remains a need for a thorough, quantitative evaluation of the \( R_s-\eta \) relationship. To direct future efforts, it is important to understand how additional OPV \( R_s \) and mobility improvements will affect \( \eta \). Previous studies provide useful guidance for modeling efficiency optimization, but their focus was primarily on energy level parameters for donor and acceptor materials. A thorough analysis of OPV efficiency optimization based on series resistance considerations is also crucial, particularly given the attention it has received in OPV research. Providing such an optimization analysis should help guide future OPV materials development. We proceed here from an established relationship describing \( J-V \) behavior in solar cells and directly accounting for resistance (\( R_s \)) effects on cell performance:

\[
J = J_0 \left[ \exp \left( \frac{e(V - \frac{1}{2}R_s)}{nk_B T} \right) - 1 \right] + \frac{V - \frac{1}{2}R_s}{R_p} - J_L \tag{3}
\]

where \( J_0 \) is the reverse saturation current, \( e \) is the elementary charge, \( V \) is the cell voltage, \( J \) is the current density, \( n \) is the diode ideality factor, \( k_B \) is Boltzmann's constant, \( T \) is temperature, \( R_s \) is the equivalent circuit shunt, or parallel resistance, and \( J_L \) is the photocurrent generated by the cell before recombination losses. The application of Equation 3 to OPVs has been previously discussed. Equation 3 stems from the original work of Shockley and co-workers to describe the ideal diode behavior of a \( p-n \) junction. Later, Shockley, Read, and Hall modified this \( p-n \) junction analysis to account for deviations from ideal diode behavior (i.e., non-ideal \( p-n \) junctions having \( n > 1 \)). OPVs exhibit the circuit characteristics expressed in Equation 3, and research is still in progress to understand the factors underlying \( J_0 \) and \( n \) in OPVs. Extensions to Equation 3 have also been made to account for the effects of light intensity. Other useful modeling approaches have also been suggested. Our approach builds on Equation 3, which enables direct and quantitative evaluation of \( R_s \) effects on cell response.

We report a comprehensive analysis of \( R_s \) effects on OPV power conversion efficiency through a combined experimental data-computational approach. We begin by studying a BHJ OPV cell design based on poly(3-hexylthiophene) and 6,6-phenyl C61-butyric acid methyl ester (P3HT:PCBM) as the baseline. P3HT:PCBM systems have shown some of the largest reported single layer cell efficiencies for OPVs and are of interest when analyzing BHJ OPV efficiency losses. The design and fabrication details for these cells were recently reported. Figure 1 shows the architecture of these cells. The devices have a nickel oxide (NiO) interfacial layer on the indium tin oxide (ITO) anode and a lithium fluoride (LiF) interfacial layer on the aluminum cathode, giving the overall cell structure glass/ITO/NiO/P3HT:PCBM/LiF/Al (note that we refer to this cell design as the “P3HT:PCBM test cell” throughout this paper). To evaluate the effects of optimizing \( R_s \), we use a least-squares fit to extract \( R_s \), as well as the other parameters in Equation 3 \((R_p, n, \text{and } J_L)\), from the previously reported current-voltage dark data. Despite the relatively low mobilities of the organic active layer materials, this analysis will show that this P3HT:PCBM cell design is approaching the theoretical limit in \( R_s \); further optimizing \( R_s \) (i.e., \( R_s = 0 \)) would lead to an improvement in \( \eta \) of only ~0.1% (from 5.2% to 5.3%). Next, since the goal of OPV development is ultimately to move beyond systems in the 5% efficiency regime, we evaluate the effects of losses due to \( R_s \) for the important scenario of projected high-efficiency OPV cells (i.e., \( \eta > 10\% \)). Finally, noting that current laboratory-scale OPV cells reported in the literature typically have modest areas (e.g., 0.06 cm\(^2\)), we evaluate the effects of series resistance losses for larger area cells, finding that the anode resistance is the only remaining materials property that significantly effects a change in \( R_s \) for P3HT:PCBM cells, as well as for other OPVs with comparable \( R_s \) values. We conclude by using this analysis to offer a framework for investigating future OPV design strategies.

2. Results

2.1. Evaluation of Simulation Accuracy

Figure 2 shows a comparison of our simulated data and previously reported experimental data, using the standard solar spectrum.
2.2. $R_s$ Effects on $\eta, J_{sc}$ and Fill Factor

Figure 3 shows simulations of how the cell $J$–$V$ characteristics change with increasing $R_s$ values. These curves are generated from Equation 3 and the parameters in Table 1, with $R_s$ the only parameter being varied. The simulation shows how $R_s$ impacts the key performance parameters: fill factor is significantly affected by increases in the P3HT:PCBM cell $R_s$, $V_{oc}$ does not change, and $J_{sc}$ only changes at very large $R_s$ values (between 10 and 100 $\Omega$ cm$^2$). Figure 3 shows that completely eliminating $R_s$ losses affords a very minor enhancement in $\eta$ of $\sim0.1$%—from 5.2% to 5.3%. Comparable experimental $R_s$ effects on the cell $J$–$V$ response of other BHJ OPVs have been reported, demonstrating the predictive value of this modeling approach in analyzing $R_s$ effects. For example, Ma et al. showed how changes in annealing conditions led to improved active layer crystallinity and therefore reduced $R_s$ values (due to increased mobilities of the more crystalline component materials).\[11\] The $R_s$ variations for their experimental samples exhibit $J$–$V$ response effects comparable to the calculated results in Figure 3. However, the lower limit of their $R_s$ values was $\sim10$ $\Omega$ cm$^2$; the present computational method can address efficiency enhancements in the limit of $R_s \approx 0$—for example, to assess how further mobility enhancements would affect efficiency losses from cell resistance.

Figure 4 shows how $\eta$ and fill factor are affected by $R_s$ variations. Since the $R_s$ value (1.4 $\Omega$ cm$^2$) for the PCBM:P3HT test cell is at the

<table>
<thead>
<tr>
<th>$R_s$ $\Omega$ cm$^2$</th>
<th>$J_{sc}$ $\mu$A cm$^{-2}$</th>
<th>$n$</th>
<th>$J_{sc}$ $\mu$A cm$^{-2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.4</td>
<td>3960</td>
<td>2.38</td>
<td>$3.65 \times 10^{-7}$</td>
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</table>

Figure 3. The effect of $R_s$ variation on simulated $J$–$V$ characteristics for the P3HT:PCBM test cell. These data assume that $R_s$ is the only parameter that changes; all other parameters in Equation 3 are held constant (i.e., $J_s, R_p, n$, and $J_{sc}$). The parameters for these curves are given in Table 1. For the test cell, $R_s = 1.4 \Omega$ cm$^2$, and $\eta = 5.2\%$. For a zero resistance cell (i.e., $R_s = 0$), $\eta$ increases by only 0.1% to 5.3%. Large $R_s$ values ($\sim100 \Omega$ cm$^2$) have significant effects on cell efficiency (e.g., an $R_s$ of 100 $\Omega$ cm$^2$ reduces the cell efficiency to 1.0%).
knee in both curves, further reductions in \( R_s \) afford only minor enhancements in cell performance, although moderate increases in \( R_s \) produce significant declines in both \( J_{sc} \) and fill factor. Note that \( \eta \) and fill factor track each other almost exactly until \( R_s \) approaches \( \sim 50 \, \Omega \, \text{cm}^2 \). At this point, losses in both fill factor and \( J_{sc} \) affect \( \eta \), and fill factor and \( J_{sc} \) diverge as shown in Figure 4. This divergence is due to effects on \( J_{sc} \) as \( R_s \) approaches \( \sim 50 \, \Omega \, \text{cm}^2 \); the \( R_s-J_{sc} \) relationship is shown in Figure 5. For the present PCBM:P3HT cell design, there is a strong \( R_s-J_{sc} \) correlation only at high \( R_s \) values; \( R_s \) must increase by almost two orders of magnitude before there is a noticeable effect on \( J_{sc} \) (at which point \( J_{sc} \) declines rapidly with further \( R_s \) increases). The same fill factor data from Figure 4 are shown in Figure 5 for comparison, highlighting the fact that in the PCBM:P3HT test cell regime (\( R_s \approx 1 \, \Omega \, \text{cm}^2 \)), fill factor is dramatically more sensitive to \( R_s \) than is \( J_{sc} \).

See Section 3 for a discussion of this analysis and the possibility of enhancements in cell performance, although moderate increases in \( R_s \) are more significant in a high-efficiency scenario, but remain modest for the test cell \( R_s \) (1.4 \( \Omega \, \text{cm}^2 \), this gives \( \eta = 13.1\% \), compared with \( \eta = 13.6\% \) for the zero \( R_s \) loss case. Note that the losses from \( R_s \) increases are significantly greater for the high-efficiency \( J-V \) curves than for the P3HT:PCBM test cell case (Fig. 3).

2.3. Evaluation of \( R_s \) Losses for a Projected High-Efficiency OPV Scenario

We evaluate here the effects of \( R_s \) losses for a scenario of high-efficiency OPV cells. To generate data for the high-efficiency case, we choose a model based upon a combination of the P3HT:PCBM test cell attributes and future target properties for OPVs.\(^{[15,20,57–59]}\) These targets address the limitations of the P3HT:PCBM and other current OPV systems, which have been previously discussed.\(^{[2,19,40]}\) The limitations reflect mismatches between donor and acceptor energy levels and the need for lower bandgap (~1.5 \( \text{eV} \)) materials. Note that the low between the lowest unoccupied molecular orbitals of the donor (LUMO\(_D\)) and acceptor (LUMO\(_A\)) materials is critical. We assume here that the optimal case is a LUMO\(_D\)/LUMO\(_A\) offset of 0.4 \( \text{eV} \). With all other factors being equal, this offset would be a significant advance over current state-of-the-art OPV systems, and it appears to be realizable judging from recent experimental\(^{[22]}\) and computational results.\(^{[39,60]}\) Some materials are expected to require significantly larger LUMO\(_D\)/LUMO\(_A\) offsets for efficient exciton dissociation (e.g., in the case of low dielectric constant environments or low carrier mobilities).\(^{[61–64]}\) Our modeling here is based upon the expectation that, at low LUMO offsets, these exciton dissociation challenges will be overcome and that other materials and device characteristics will remain on par with the present P3HT:PCBM system (i.e., equivalent \( R_s, R_p, n, \) and external quantum efficiency (EQE) values). These energy level optimizations would allow for increased \( J_{sc} \) through increased light absorption (i.e., lower bandgap materials) and increased \( V_{OC} \) through better donor-acceptor energy level matching.\(^{[2,19,40]}\) See Ref. \([65]\) for further details on this simulated high-efficiency case.

Figure 6 provides the results for a projected high-efficiency scenario with \( R_s = 1.4 \, \Omega \, \text{cm}^2 \) (the experimental value for the PCBM:P3HT test cell), with \( R_p = 0 \) and larger \( R_s \) values. A summary of the input parameters is given in Table 2.\(^{[66]}\) The \( R_s \) optimization in Figure 6 shows that the effects of an optimized \( R_s \) are more significant in a high-efficiency scenario, but remain modest for the test cell \( R_s \) (1.4 \( \Omega \, \text{cm}^2 \)). For \( R_p = 1.4 \, \Omega \, \text{cm}^2 \), \( \eta = 13.1\% \), and for \( R_s = 0, \eta = 13.6\% \) – giving a 0.5% efficiency loss for an \( R_s \) of 1.4 \( \Omega \, \text{cm}^2 \), versus 0.1% loss in the P3HT:PCBM test cell case. The efficiencies for \( R_s = 0 \) are calculated from Equation 3 and by applying the parameters in Tables 1 and 2. The greater sensitivity to \( R_s \) in this high-efficiency scenario is because the high-efficiency cell has a greater current density, as shown in Figure 6. Efficiency losses due to series resistance can be approximated as \( \sim J_{sc}^{\text{max}} R_s \).\(^{[49]}\) In this high-efficiency scenario, the operating current

<table>
<thead>
<tr>
<th>( R_s ) [( \Omega , \text{cm}^2 )]</th>
<th>( J_{sc} ) [( \text{mA cm}^{-2} )]</th>
<th>( n )</th>
<th>( J_L ) [( \text{A cm}^{-2} )]</th>
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</tr>
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<tr>
<td>1.4</td>
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<td>3.45 x 10^{-8}</td>
<td>0.0224</td>
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is approximately twice that of the P3HT:PCBM test cell (Figs. 3 and 6). It would be expected, therefore, that doubling this current density will increase resistive losses by approximately 4x. This is the case; efficiency losses due to the $R_s$ increase from ~0.1% for the P3HT:PCBM test cell to ~0.5% in this high-efficiency scenario.

2.4. Effects of Cell Area on $R_s$

The above analysis of the P3HT:PCBM test cell shows near-optimal behavior in $R_s$. However, the cell area is only 0.06 cm$^2$ — typical of laboratory-scale OPVs — and it is therefore necessary to consider how losses due to $R_s$ change with increases in cell area. Gupta et al. studied cell area effects on power conversion efficiency, although this study primarily focuses on illumination effects from flooding the cell at its perimeter (with additional current generated at the perimeter due to scattering effects).[67] Xue et al. evaluated changes for varying experimental cell areas, but only up to 0.055 cm$^2$. [10]

When considering the effect of cell area on $R_s$, the transparent anode resistance is the dominant factor. The aluminum cathode contributes negligible resistance compared to the ITO anode, given the conductivity ratio of ~100.[68,69] Furthermore, the active layer resistance contribution does not increase with increasing cell area. Figure 7a shows this graphically for hole transport (the same analysis applies for electrons). Regardless of cell area, charge carriers travel the same distance (perpendicularly) through the active and interfacial layers, and thus, the losses due to the resistance of these layers are independent of cell area. In contrast, losses due to anode resistance increase with cell area because of the lateral flow of charge through the anode. The following simple relation is a useful way to analyze anode contributions to $R_s$:

$$R_s = R_{s,1} + R_{s,\text{anode}}$$  \hspace{1cm} (4)

where $R_{s,1}$ contains the contributions from the active layer, interfacial layers, current collectors, and interconnects, and $R_{s,\text{anode}}$ is the contribution to $R_s$ from the transparent anode. Figure 7b illustrates Equation 4 with a simple diagram, where $R_s$ is divided into these two primary resistors in series. Note that $R_{s,1}$ is independent of cell area, whereas $R_{s,\text{anode}}$ is a function of cell area.

The relationship for this resistance contribution is determined to be:

$$R_{s,\text{anode}} = \frac{w^2}{3\sigma_{\text{anode}}h_{\text{anode}}} + \frac{ww'}{\sigma_{\text{anode}}h_{\text{anode}}}$$ \hspace{1cm} (5)

where $\sigma_{\text{anode}}$ is the anode conductivity, $w$ is the cell active layer width, $h_{\text{anode}}$ is the anode thickness, and $w'$ is any additional distance between the cell perimeter and the current collector (i.e., any additional distance over which the current must flow through the ITO before reaching the current collector); a related effect is reported for inorganic photovoltaic cells.[70] For reference, we derive Equation 5 in the Supporting Information for this paper. When $w'$ is negligible, the second term in Equation 5 goes to zero. We assume square cell geometries in determining cell area (i.e., cell area = $w^2$). Equation 5 enables one to calculate differences in overall $R_s$ based on changes in cell area and anode conductivity. Applying Equations 4 and 5, we determine $R_{s,\text{anode}} = 0.5 \, \Omega \cdot \text{cm}^2$ and $R_{s,1} = 0.9 \, \Omega \cdot \text{cm}^2$ for our P3HT:PCBM test cell. Therefore, as cell area increases, $R_{s,\text{anode}}$ will increase according to Equation 5; however, $R_{s,1}$ will remain constant at 0.9 $\Omega \cdot \text{cm}^2$ regardless of cell size.

Figure 8 illustrates the factors that underlie the resistive losses arising from the anode: the anode conductivity and the cell area. For the details of the computational methodology for this analysis, see Section 5. The analysis assumes that the anode thickness remains constant.[71] Figure 8 shows that for cell areas under ~0.2 cm$^2$, even at relatively low anode conductivities, the efficiency loss due to anode resistance is minor. However, efficiency losses for larger cells (>0.2 cm$^2$) can clearly be substantial. For these larger cell dimensions, the importance of the anode conductivity increases significantly, as indicated by efficiency losses shown on the right side of the graph. When this loss analysis due to anode resistance is applied to the projected high-efficiency scenario (Section 2.3), there is even greater sensitivity to anode conductivity and cell size; Figure 9 shows these results. For low anode conductivities, efficiency losses due to $R_s$ become significant at areas greater than ~0.02 cm$^2$ (compared with 0.2 cm$^2$ for the test cell case). The greater sensitivity of efficiency losses to cell area and anode conductivity is also evident in the tighter packing of the contours in Figure 9, compared with the results for the P3HT:PCBM test cell in Figure 8.
...and charge carrier concentration. Conductivity is the resistance of a material is affected by three factors – mobility, cell dimensions, and charge carrier concentration. Conductivity is calculated by:

\[ \sigma = en\mu \]  

where \( n \) is the charge carrier density and \( \mu \) is the carrier mobility. One can therefore calculate the resistance of a material from its conductivity (Equation 6) and the cell dimensions. The \( R_s \) contribution from the active layer has been measured experimentally, where increasing the active layer thickness (\( h_{AL} \)) gives a linear response in \( R_s \):

\[ \Delta R_{S,AL} \propto \frac{\Delta h_{AL}}{\sigma_{AL}} \]  

(7)

where \( R_{s,AL} \) is the active layer contribution to \( R_s \), and \( \sigma_{AL} \) is the active layer conductivity. Equations 6 and 7 show that mobility is only one aspect of the active layer contribution to the cell resistance, and even for relatively low mobilities, efficiency losses due to \( R_s \) can be minimized. In comparing Equations 5 and 7, note that the \( h \) term appears in the denominator in Equation 5 and in the numerator in Equation 7. Intuitively, this is because charge carriers effectively make a right-angle turn at the electrodes, as shown in Figure 7a. Consequently, a larger \( h_{AL} \) means that charge carriers must travel further through the active layer and incur greater resistive losses, whereas a larger \( R_{s,anode} \) means that they have a wider (and thus, lower resistance) pathway through the anode.

The present results show that, while recent advances in P3HT:PCBM mobilities have likely led to significant efficiency enhancements via reductions in the active layer \( R_s \) contributions, further P3HT:PCBM active layer mobility enhancements will do little to overcome resistance losses since the performance of this particular cell is already approaching optimal behavior in \( R_s \). For example, the annealing and self-organization experiments on P3HT:PCBM reported by Li et al. improved active layer hole mobilities by a factor of \( \sim 10 \) and reduced \( R_{s} \) values by a factor of \( \sim 10x \) (from 19.8 \( \Omega \) cm\(^2\) to 2.4 \( \Omega \) cm\(^2\)).[36] Furthermore, \( \eta \) of the high mobility devices was \( \sim 4\% \), versus \( \sim 1\% \) for the lower mobility devices. The analysis in Figure 4 demonstrates that a substantial portion of this efficiency enhancement is due to \( R_s \) reduction. However, Figure 4 also indicates that further reductions in \( R_s \) below \( \sim 1\ \Omega \) cm\(^2\) would not significantly enhance \( \eta \).

It is also important to consider potential coupling between \( R_s \) and other cell characteristics. With increasing \( R_s \), the approach used here yields significant losses to fill factor, losses to \( J_L \) when \( R_s \) is sufficiently high (\( \sim 50 \Omega \) cm\(^2\)), and no changes to \( V_{oc} \) (Figs. 3–6). By Ohm’s Law, a change in voltage \( \Delta V = J \Delta R \). However, when \( J = 0 \) (i.e., at \( V = V_{oc} \)), \( \Delta V = 0 \) as well, and therefore \( V_{oc} \) is not expected to change, as shown here. Experimentally, this result has also been observed where changes to \( R_s \) yield almost no change to \( V_{oc} \).[6,11,30] One area where \( R_s \) may be significantly coupled to another parameter is \( J_L \), due to mobility effects, thereby affecting \( J_L \). While \( R_s \) and mobility are closely linked via Equations 5, 6, and 7, mobility certainly affects other performance parameters in a solar cell beyond \( R_s \). A large \( R_s \) is sometimes a result of low mobility materials (Equations 6 and 7), and previous reports show that low mobilities can reduce \( J_L \) if charge carrier lifetimes are less than their respective collection times.[42,54] This effect manifests itself in the \( J-V \) curve through a non-zero slope near short circuit and a reduction in \( J_{sc} \). The effect is negligible for the present P3HT:PCBM test cell, however, as evidenced by the high quantum efficiencies and near-zero-slope \( J-V \) behavior at a zero cell voltage (Fig. 2).[14] That said, we expect that \( \eta \) is more sensitive to mobility than to \( R_s \). For example, while a 10x increase in \( R_s \) has a negligible...
effect on $J_{sc}$ (see Fig. 5), the analysis reported by Schultinsky et al. indicates that a $10 \times$ increase in electron and hole mobilities would have an appreciable deleterious effect on $J_{sc}$ and thus $J_{sc}$; see their discussion for details on this proposed mobility–$J_{sc}$ relationship.\[12\] Ultimately, however, this point simply highlights our assertion that state-of-the-art P3HT:PCBM mobilities appear to be sufficient for overcoming most losses due to resistance and charge collection. Other areas where mobility is relevant are the various conduction processes in semiconductors, such as charge injection through metal-semiconductor (Schottky) barriers\[26,74] and Poole–Frenkel emission.\[17,75\] Furthermore, while research is ongoing in understanding exciton dissociation dynamics in OPVs\[60–64,76–78\] it is possible that greater carrier mobilities could enhance exciton dissociation rates, which play a crucial role in improving overall OPV power conversion efficiencies.\[2,4,61,64]\]

Finally, the one area where there is immediate opportunity for $R_p$ optimization in state-of-the-art OPVs is in the anode conductivity for larger cell sizes. Figures 8 and 9 demonstrate this conclusion; for cell areas greater than $\sim 0.2 \text{cm}^2$ and $\sim 0.02 \text{cm}^2$, respectively, anode conductivity becomes increasingly important in reducing losses due to cell resistance. Alternative transparent anode materials have received significant attention in recent years,\[79–83\] and transparent conducting oxides (TCOs) with exceptionally high conductivities of over 17,000 S cm$^{-1}$ have been reported.\[68,84\] These high conductivity TCOs are approaching the intrinsic limit of $\sim 25,000 \text{S cm}^{-1}$ proposed by Bellingham, Phillips, and Adkins.\[85\] Figures 8 and 9 show the cell sizes at which these high conductivities are important in OPVs: if an OPV manufacturer sought to reduce costs through increasing current collector spacing (i.e., larger cell areas), then these high conductivity TCO materials would become increasingly important.

### 4. Conclusions

We show here that efficiency losses arising from cell resistance ($R_p$) in optimized glass/ITO/NiO/P3HT:PCBM/LiF/Al BHJ solar cells are minor: removing all losses due to cell resistance is estimated to lead to an efficiency improvement of only $\sim 0.1\%$ (from $\eta = 5.2\%$ to $\eta = 5.3\%$) in our test cell. Resistive losses become significant only for relatively large cell dimensions, where anode conductivity is an important contributor to these losses. For a projected future high-efficiency OPV scenario, losses due to $R_p$ become more significant, but remain modest for our test cell area of 0.06 cm$^2$ (increasing to 0.5% vs. 0.1% for the present cells). Furthermore, there is greater sensitivity to cell size for cell areas greater than $\sim 0.02 \text{cm}^2$ in projected high-efficiency OPVs. Therefore, for future high-efficiency OPVs, a key component is likely to be higher conductivity anode materials, which would substantially reduce $R_p$ losses for modest cell sizes and are the only means to impact $R_p$ significantly in these cells (assuming an active layer contribution to $R_p$ comparable to P3HT:PCBM). The resistance of the P3HT:PCBM active layer, on the other hand, is nearly optimal; with all other factors remaining equal, further mobility improvements in these active layer materials should have minor effects in reducing resistance losses. Further studies will be necessary to articulate more completely how high mobility OPV materials affect other types of losses (e.g., charge carrier recombination or exciton dissociation losses).

It is evident that OPVs will not advance beyond the 10% efficiency threshold without new donor and acceptor materials that increase $V_{oc}$ and absorb larger portions of the solar spectrum for enhanced $J_{sc}$ values. Developing modeling and analytical approaches is crucial to understanding the requirements for these new materials, particularly regarding the question of required LUMO$_D$–LUMO$_A$ separation. For future OPV research efforts, resistance losses are therefore always important considerations, but new active layer materials are critical to overcoming $V_{oc}$ and $J_{sc}$ losses incurred in today’s best OPV systems.

### 5. Computational Methodology

To determine the dark fit parameters, a least-squares fit was employed to solve numerically for the parameters, applying Equation 3 without the $J_0$ term (as there is no photocurrent in the dark). Using these fit parameters, it was then possible to apply Equation 3 in simulating $J$–$V$ data based on changes in $R_p$, equating the experimental $J_{sc}$ to $J_{sc}$ according to Ref.\[28\] Waldauf et al. have shown how $R_p$ varies with light intensity (see Ref. [49]). To address this dependence, the same method used for the dark data was applied to the light data and an $R_p$ of 3960 $\Omega$ cm$^2$ for the light $J$–$V$ curve was determined; the dark $R_p$, on the other hand, was determined to be 41,400 $\Omega$ cm$^2$. While the difference between the light and dark $R_p$ values is large, this difference has only a slight effect on the behavior of the simulated curves for this data set, and both light and dark $R_p$ values therefore give excellent simulations of the cell response. To generate Figures 4 and 5, which show the effects of $R_p$ on $J_{sc}$, fill factor, and $\eta$, Equation 3 was solved numerically to generate new $J$–$V$ data sets for successive changes in $R_p$ and to calculate $J_{sc}$, fill factor, and $\eta$. To calculate losses due to anode resistance (Figs. 8 and 9), Equation 3 was solved numerically to generate new $J$–$V$ data sets at each point and to determine new maximum $\eta$ values. This process requires first extracting the P3HT:PCBM test cell anode contribution to $R_p$ to calculate a "zero loss" case and then determining the new anode $R_p$ contribution based upon modeled anode conductivity and cell area.

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Note that while increasing the anode thickness will decrease the anode contribution to the optical transmission of the anode is reduced with increasing thickness, there is a counter effect in that the optical transmission of the anode is reduced with increasing thickness. Therefore, the best way to address the anode contribution to Rs is to increase the anode thickness to reduce its optical transmission. This counter effect is well-established for the high-efficiency, high-efficiency solar cells (see Refs. [45,46]), it is not well-developed for BHJ OPVs, and studies are still in progress. Discussion of the parameter can be found in Ref. [41].

Note that while increasing the anode thickness will decrease the anode contribution to Rs, as shown in Equation 5, there is a counter effect in that the optical transmission of the anode is reduced with increasing thicknesses. Therefore, the best way to address the anode contribution to Rs is to use higher conductivity materials.

Note that charge generation profiles can also affect how an increase in active layer thickness affects Rs. For example, if more electrons form near the transparent anode, they have a larger distance to travel before arriving at the aluminum cathode (thus affecting resistance losses), compared with forming near the cathode. This charge generation profile will be system-specific; see Ref. [4] for BHJ OPV profiles.