

Response to Christopher P. Muzzillo's Comments on "Introduction of a Novel Figure of Merit for the Assessment of Transparent Conductive Electrodes in Photovoltaics: Exact and Approximate Form"

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Similarities and differences between figure of merits (FOMs) for the assessment of transparent conductive electrodes (TCEs) are discussed. This article is a response to C. P. Muzzillo's comment on the introduction of the novel FOM (the so-called exact FOM or Anand's FOM) and it deals with questions about how implicit and how exact the different approaches really are and whether specific application cases can be covered or not. While the exact FOM has been introduced to provide an upper limit of photovoltaic power conversion efficiency for the whole range of possible transmittance and sheet resistance values of transparent conductive oxides, Muzzillo's comment points out specific application cases, that have to be treated with more individual modeling. In this work, the authors adopt these application cases into the exact FOM to demonstrate its applicability. Furthermore, the FOM approximation given by Muzzillo is used and slightly refined, yielding an even better agreement with the exact FOM. In the end, it is concluded that both approaches are justified: Muzzillo's FOM for very practical applications and Anand's (exact) FOM for fundamental assessment. In this work, both approaches have been harmonized to yield an ultimate tool for the future development of TCEs for photovoltaics.

1. Introduction

Over the past decades, several more or less physically justified assessments of transparent conductive electrodes (TCEs) for use in photovoltaics have been presented in terms of the figure of merits (FOMs).^[1–7] Our ultimate target with the article

mentioned in the title was to establish a novel FOM, that would as by demand fulfill the following properties: i) Being proportional to the potential power output of photovoltaic devices, ii) being normalized with regards to the theoretical maximum of photovoltaic performance, and finally, iii) being a guide for developing novel TCEs.^[7] This could be straight-forward accomplished by using detailed-balance numerical calculations considering the Shockley–Queisser Limit (SQL) as maximum photovoltaic power output. However, solving the Shockley equation this required application of numerical methods, which are not typically used for computing FOMs in general. This point has been correctly raised by C.P. Muzzillo in his comment on our article alongside two other aspects, namely the extension of the model towards application for various solar module geometries and the impact of adding current-collecting grids. As we learned from the comment of Muzzillo,

there have already been some explicit assessments done for predicting the potential solar cell performance as a function of the TCE properties,^[8,9] which were, however, restricted to the special case of using additional metal grids. The most original reference done by Muzzillo was referring to a work by M.A. Green,^[10] which did neither contain a "figure of merit" nor dealt with TCEs and therefore was not recognized by us. Analyzing all that, we note the explicit FOM mentioned by Muzzillo in his comment to be shown there for the first time for general use (without metal grids) and thus this provides clear justification for his comment. While we appreciate Muzzillo's sense for computing details of special cases, our original intention was restricted to provide an absolute upper limit for potential photovoltaic performance and thereby disregarding any impact of independent contributions e.g., due to parallel or contact resistances. Nevertheless, we here took up the stimulation by Muzzillo to extend our model for a variety of situations in order to demonstrate its general applicability. We are intended to provide the combined benefits to users worldwide in form of an online tool, that is going to compute the FOM for their TCE and their intended solar module geometry, eventually supporting their research and development. Furthermore, we do agree with Muzzillo that specifically for general calculations of

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any solar module geometry, ultimately network simulations are required, which are—in contrast to the one-diode model used here—capable of correctly considering the impact of distributed series resistances on power conversion efficiency.^[11] However, such simulation is beyond the scope of this response and might be considered in future work. We shall however mention that we have investigated the differences between the one-diode model with a single lumped series resistance and the micro-diode model (standing for all network simulations) with a distributed series resistance.^[11] Within a confidence range around the optimal solar cell length (0.3–0.5 cm), we found the error done by lumping the series resistance in the one-diode model to be very small (<1%) for sheet resistances up to 100 Ω square⁻¹. As consequence, the “exact FOM” as presented before provides indeed very little error with regard to a network model within the constraints of having near optimal solar cell lengths. Finally, we still consider the “exact FOM” as appropriate naming, since the impact of each physical parameter of the TCE, i.e., the optical transmittance and the sheet resistance (or simply electrical conductance) is being considered in that model directly and physically “as is”, without any filtering or estimation.

2. Approximations, Deviations, and the Question of Explicitness

Based on the approximation, that the TCE impact on the maximum power point current density J_{MP} and the maximum power point voltage V_{MP} is solely arising from the limited transmittance and series resistance losses, Muzzillo gets:

$$J_{MP,TCE} = J_{MP} \cdot T_{TCE,avg} \quad (1)$$

$$V_{MP,TCE} = V_{MP} - R_{TCE} \cdot A \cdot J_{MP} \quad (2)$$

Consequently, Muzzillo defines his figure of merit as:

$$\phi_{Muzzillo} = \frac{J_{MP,TCE} \cdot V_{MP,TCE}}{J_{MP} \cdot V_{MP}} = T_{TCE,avg} \cdot \left(1 - \frac{R_{sh,TCE} \cdot L^2 \cdot J_{MP}}{3 \cdot V_{MP}} \right) \quad (3)$$

In fact, Muzzillo’s FOM is based on a few earlier works as indicated by himself.^[12] Although Muzzillo’s figure of merit (Equation 3) is claimed to be an explicit FOM, the calculation of current density and voltage at the maximum power point requires solving the Shockley equation as well, which is the very implicit equation we are using for our exact FOM. Thus, to verify $\phi_{Muzzillo}$, we numerically solved the implicit Shockley equation to derive the current-voltage curve, which then was evaluated for the current density and voltage at the maximum powerpoint. However, if in practical cases real solar cells are considered, the maximum power point parameters could be computed and evaluated from the measured IV characteristics, making in such case the FOM provided by Muzzillo indeed an explicit approach. Muzzillo mentioned his figure of merit to be a good approximation to our exact FOM (or ϕ_{Anand}) in case there were no module geometries or additional metal grids to be considered.^[12] Looking at his formulas and the involved approximations we found a simple way to approach the exact FOM even closer by simply considering the limitation of the photocurrent

by the TCE (compare with equation 1) also for the correction done on the maximum power point voltage (compare with Equation 2):

$$V_{MP,TCE} = V_{MP} - R_{TCE} \cdot A \cdot J_{MP} \cdot T_{TCE,avg} = V_{MP} - \frac{R_{sh,TCE} \cdot L \cdot J_{MP} \cdot T_{TCE,avg}}{3} \quad (4)$$

And thus, the modified Muzzillo figure of merit readily becomes:

$$\phi_{Muzzillo,mod} = \frac{J_{MP,TCE} \cdot V_{MP,TCE}}{J_{MP} \cdot V_{MP}} = T_{TCE,avg} \cdot \left(1 - \frac{R_{sh,TCE} \cdot L^2 \cdot J_{MP} \cdot T_{TCE,avg}}{3 \cdot V_{MP}} \right) \quad (5)$$

Upon comparison (see **Figure 1**) between $\phi_{Muzzillo}$, $\phi_{Muzzillo,mod}$ and ϕ_{Anand} (the “exact FOM”), we discover a remarkable improvement for $\phi_{Muzzillo,mod}$ within the range between 1 and 100 Ω square⁻¹, reducing there the difference to ϕ_{Anand} to below 1% (Figure 1 subset). Above 100 Ω square⁻¹ both approximations quickly degrade to an absolute error exceeding 10% with acceleration.

Nevertheless, for sheet resistances surpassing about 200 Ω square⁻¹, both approximations ($\phi_{Muzzillo}$ and $\phi_{Muzzillo,mod}$) deviate strongly and even turn negative. While that range of sheet resistances is not reasonable for any photovoltaic development, it indicates the limitations of the model shown by Muzzillo.

Intrigued by Muzzillo’s FOM and in order to put it in a broader perspective, we reassessed the figure of merits (FOMs) in the same way, as in our original article. Overlooked before, we also added the FOM published by Jain and Kulshreshtha in 1981,^[3] which is based on a simple ratio of the optical absorption coefficient (α) and the electrical conductivity (σ). Our general assessment does not change based on the current extensions, since the “exact FOM” or “ ϕ_{Anand} ” remains the only FOM, that is capable of quantitatively describing the impact of the sheet resistance over the wide range shown here in **Figure 2**.

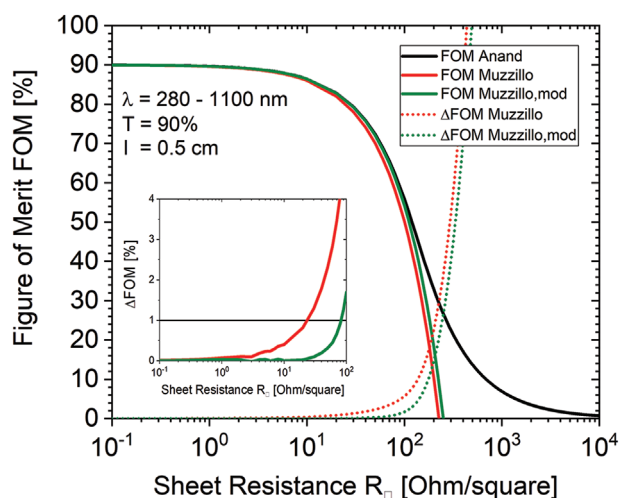


Figure 1. Comparison between ϕ_{Anand} (the exact FOM), $\phi_{Muzzillo}$, $\phi_{Muzzillo,mod}$ for the wavelength range of 280 to 1100 nm, a constant transmittance of 90%, and a solar cell length of 0.5 cm. For the calculation of $\phi_{Muzzillo}$ and $\phi_{Muzzillo,mod}$, the $J_{MP,TCE}$ was 42.15 mA cm⁻² and $V_{MP,TCE}$ was 793.9 mV—taken from Muzzillo’s comment.

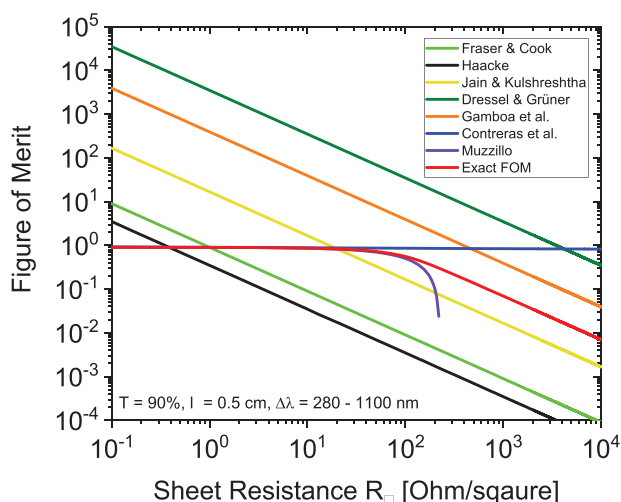


Figure 2. Comparison between different figures of merit for the wavelength range of 280–1100 nm, a transmittance of 90% over the wavelength range, and a solar cell length of 0.5 cm.

The double logarithmic plot shows that FOMs by Fraser & Cook, Haacke, Jain & Kulshreshtha, Dressel & Grüner, and Gamboa et al. (Figure 2) exhibit the same behavior. For an equal change in sheet resistance, they span a five-order-of-magnitude range but are separated from each other by specific prefactors. This is not surprising, given that they're all proportional to the inverse of the sheet resistance ($1/R_{\square}$) and just differ in the constant proportionality factor. In contrast, the FOM by Contreras et al. ($n = 100$) varies by less than one order of magnitude and has a very moderate dependence on sheet resistance, pointing out a specific realism that has not been obtained before. Muzzillo's FOM initially follows very closely the exact FOM up to about $100 \Omega \text{ square}^{-1}$, but then rapidly turns negative not far above $200 \Omega \text{ square}^{-1}$. While one may argue whether sheet resistances above $100 \Omega \text{ square}^{-1}$ may be of any practical relevance, it remains our desire to be capable of quantitatively describing the FOM for arbitrary values of the TCE's sheet resistance.

All historical FOMs exhibit an emphasis on the sheet resistance over the optical transmittance and since they are not normalized, smaller sheet resistances always win over insufficient transmittance, which may contradict the obtainable photovoltaic performance. Only the FOMs (or fill factor as called by the originators) by Muzzillo and forerunners as well as by Contreras et al. display exceptions from this rule—both approaches capture well the transmittance limited regime. However, in both cases, the FOM value in the regime for larger sheet resistances is deviating strongly from the expected performance of a solar cell. In summary, the exact FOM captures the correct balance of influences between TCE transmittance and its sheet resistance, which is also represented by a hyperbolic dependence on sheet resistance for higher values of the same.

3. The Figure of Merit for Solar Modules

In his comment, Muzzillo points out that our calculation does not consider the effects of the geometric fill factor (GFF)

of solar modules. As indicated in our original work, we were intended to only consider effects directly arising from the physical properties of the TCE itself, namely its transmittance and its sheet resistance. However, as shown by Muzzillo and us before,^[12,13] the impact of the geometric fill factor is readily added by a prefactor considering the ratio between the solar cell length over the sum of the solar cell length and the solar cell distance (dead space). Following his suggestion, we simply added this prefactor for comparison by considering the serial interconnection due to some finite solar cell distance. Thus, ϕ_{Anand} for monolithic solar modules becomes:

$$\phi_{\text{Anand, mono}} = \frac{P_{\text{MPP,TCE}}}{P_{\text{MPP,ideal TCE}}} \cdot \frac{l}{l + \Delta l} \quad (6)$$

where l is the solar cell length and Δl is the solar cell distance (or “dead space”) between adjacent cells within the module.

Aiming to adapt the FOMs provided by Muzzillo and to analyze individual series resistance impacts on the current collection, we divided the effective series resistance for various individual contributions. **Figure 3** displays a cross-sectional and top-view structure of a monolithic solar module for further reference.

3.1. Impact of TCE Bridge on Series Resistance

To consider a more realistic scenario, we also included the effect of series resistance arising from the serial interconnection due to the TCE bridge (see Figure 3).

For a solar cell of length l and width w , assuming that the solar cell distance Δl can be sub-divided into three (P1–P3) equally wide laser trenches,^[14] the effective sheet resistance of the TCE bridge becomes:^[13]

$$R_{\text{TCE,bridge}} = R_{\square} \cdot \frac{l_{\text{bridge}}}{w_{\text{bridge}}} = R_{\square} \cdot \frac{\Delta l}{3w} \quad (7)$$

where the bridge length l_{bridge} equates to one-third of the total solar cell distance Δl . This equation is obvious since any sheet resistance for a tile of length L and width W with a constant current running over it becomes:

$$R_{\text{tile}} = R_{\square} \cdot \frac{L}{W} \quad (8)$$

However, in the case of a solar cell, the current is linearly built up in the current transport direction, and therefore the effective series resistance for the total photocurrent and due to the sheet resistance becomes upon integration:^[13]

$$R_{\text{TCE,eff}} = R_{\square} \cdot \frac{l}{3 \cdot w} \quad (9)$$

Thus, the total series resistance (R_s) due to the TCE for monoliths can be calculated as:

$$R_{s, \text{mono, bridge}} = R_{\text{TCE,eff}} + R_{\text{TCE,bridge}} \quad (10)$$

We calculated the $\phi_{\text{Anand, mono bridge}}$ by solving the Shockley equation using the series resistance as defined in Equation (10).

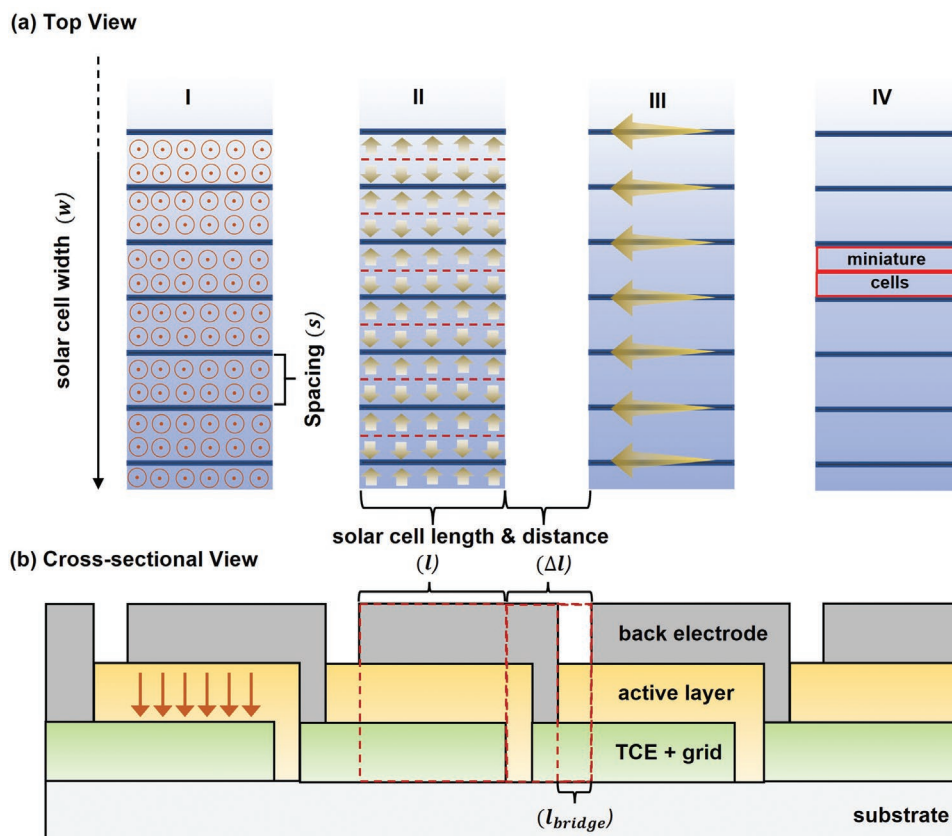


Figure 3. Schematic of a monolithic solar module with serial interconnection in the (a) top and (b) cross-sectional views. The arrows indicate the hierarchical current flow directions: I) vertically within, II) horizontally on top, and III) within the grid fingers, perpendicular to both the latter ones.

Note that in this case, we did not consider any impact due to the contact resistance located at the interconnect between the TCE and the back electrode. For including such an additional input parameter, additional experimental measurements (or simulations) would be required.

3.2. Impact of Grid Lines on Current Collection and Series Resistance

When using additional grid lines on top of or embedded into a TCE, the current collection can be hierarchically subdivided into several stages. Upon charge generation, the charges flow towards the TCE (see I in Figure 3a). Within the TCE, charges will be collected towards the grid lines, since its lower resistance results in potential gradients pointing to them within the TCE (see II in Figure 3a). The current collection within an area of $s/2$ times l , the TCE towards the grid lines can be described as:

$$R_{\text{TCE, coll}} = R_{\square} \cdot \frac{s}{2} \cdot \frac{1}{3l} \quad (11)$$

where $s/2$ corresponds to a miniature solar cell length and l being its width (see in Figure 3). Since here we considered only a fraction of a single cell (see IV in Figure 3a), namely

$$A_{\text{cell, miniature}} = A_{\text{cell}} \cdot \frac{s}{2w} \quad (12)$$

the impact of the total current from the full device area needs to be taken into account by multiplication with the same factor, effectively reducing the current considered to a fraction corresponding to the miniature cell area:

$$R_{\text{TCE, grid, eff}} = R_{\square} \cdot \frac{s}{2} \cdot \frac{1}{3l} \cdot \frac{s}{2w} = R_{\square} \cdot \frac{s^2}{12lw} \quad (13)$$

Once collected by the grid, the current finally flows in the direction of the solar cell length within the grid lines (see III in Figure 3a), again being subject to collecting a linearly increasing amount from the TCE on its way.

The series resistance due to the grid sheet resistance ($R_{\square, \text{grid}}$) can be calculated as:

$$R_{\text{grid, eff}} = R_{\square, \text{grid}} \cdot \frac{l}{3w} \quad (14)$$

since the grid collects the current from a full cell area ($l \cdot w$).

The effective sheet resistance of the grid can be calculated by using the standard formula of sheet resistance using the grid metal resistivity (ρ) and the thickness of the metal grid (t):

$$R_{\square, \text{grid}} = \frac{\rho}{t} \quad (15)$$

where the hypothetical, respectively the equivalent thickness can be calculated by considering the grid line volume over the area covered by the grid line for current collection:

$$t = \frac{\text{Volume}}{\text{Area}} = \frac{w_{\text{grid}} \cdot h \cdot l}{l \cdot (s + w_{\text{grid}})} = \frac{w_{\text{grid}} \cdot h}{(s + w_{\text{grid}})} \quad (16)$$

where w_{grid} is the gridline width, h is the height of the metal grating and t is the equivalent thickness for a homogeneous metal layer exhibiting the same volume as the grid lines. Thus, the total series resistance (R_s) becomes:

$$R_{s,\text{grid}} = R_{\text{TCE,grid,eff}} + R_{\text{grid,eff}} \quad (17)$$

The $\phi_{\text{Anand, grid}}$ was also computed by solving the Shockley equation with the series resistance defined in Equation (15). The overall transmittance due to metal grating would be reduced by the factor ($T_{\text{grid}} = \frac{s}{s+w}$) and thus the transmittance factor was multiplied to the figure of merit:

$$\phi_{\text{Anand, grid}} = \frac{P_{\text{MPP,TCE}}}{P_{\text{MPP,Ideal TCE}}} \cdot T_{\text{grid}} \quad (18)$$

3.3. Impact of Monolithic Serial Interconnection Using Grid Lines

Summarizing the effects due to the distance of the solar cell within the serial interconnection and the impact of additional grid lines yields the following figure of merit:

$$\phi_{\text{Anand, grid, mono}} = \frac{P_{\text{MPP,TCE}}}{P_{\text{MPP,Ideal TCE}}} \cdot T_{\text{grid}} \cdot \frac{l}{l + \Delta l} \quad (19)$$

Considering the additional effect of the TCE bridge on the series resistance:

$$R_{\text{grid, bridge}} = R_{\square, \text{grid}} \cdot \frac{\Delta l}{3w} \quad (20)$$

Thus, the total effective series resistance is defined as:

$$R_{s,\text{grid, mono, bridge}} = R_{\text{TCE, grid, eff}} + R_{\text{grid, eff}} + R_{\text{grid, bridge}} \quad (21)$$

By using the series resistance described in Equation (21), $\phi_{\text{Anand, grid, mono, bridge}}$ was calculated by solving the Shockley equation.

The results for all cases discussed above are shown in **Figure 4** for varying solar cell lengths. The graphs nicely reproduce the results shown by Muzzillo in his comment article. The optimal thickness for monoliths via $\phi_{\text{Anand, mono}}$ was also found to be 0.4 cm which provides evidence for the reproducibility of the results and the applicability of ϕ_{Anand} with various extensions.

The influence of the grid on the figure of merit throughout the solar cell length can be observed in Figure 4. Compared to solar cells with only transparent conducting electrodes, the grid expands the range of solar cell lengths that can be used for construction, which was already shown by C. P. Muzzillo. In addition, we show here that the series resistance due to the transport inside the TCE or TCE-grid bridge is so minimal, that it does not change the overall assessment of the FOM. Thus, we were able to show that the application of the exact figure

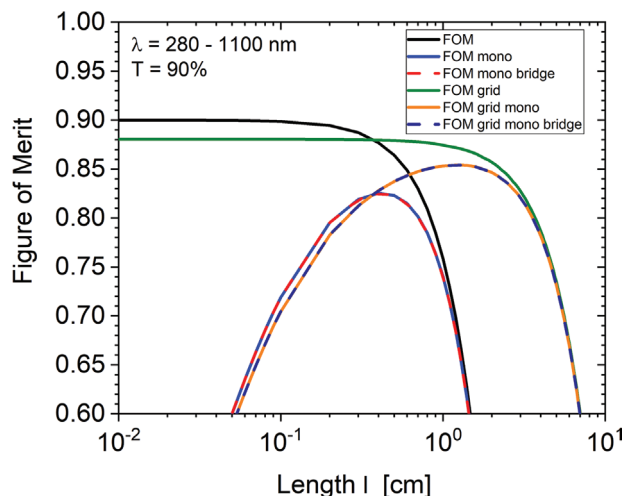


Figure 4. Figures of merit versus cell length (l): Calculations used: sheet resistance (R_{\square}) = $10 \Omega \text{ square}^{-1}$, wavelength (λ) = 280–1100 nm, Transmittance (T) = 90%, monoliths deadspace (d) = 250 μm , resistivity of grid metal (ρ_{metal}) = $10^{-5} \Omega \text{ cm}$, width of grid metal strip (w_{grid}) = 56 μm , and height of grid metal (h_{metal}) = 13 μm .

of merit can be broadened for various solar module geometries—including scenarios with additional metal grids. Overall, we were able to show that the exact FOM (or ϕ_{Anand}) could be successfully extended to cover the use-cases mentioned by Muzzillo in his comment. Nevertheless, the exact FOM in its original and simple form remains a useful upper limit concerning the attainable FOM and a lower limit concerning the overall losses, while adding an additional metal grid may yield an improvement for specific use cases. Finally, we agree with Muzzillo that further work could be directed towards the application of network simulations to properly consider the impact of distributed series resistance arising from the use of TCEs. However, this is beyond the scope of this article and maybe addressed by future studies.

4. Conclusions

Intrigued by Muzzillo's comment, we took up the challenge and extended the exact FOM (or ϕ_{Anand}) towards several use cases for calculation of the maximum power conversion efficiency factor coming along with serial cell interconnections within monolithic solar module geometries and possible extensions with added metal grids. Overall, we could reproduce all results presented by Muzzillo, using a modified version of the exact FOM.

Even though the calculational effort of the exact FOM may be larger thanks to its implicitness, we note that Muzzillo's FOM also requires numerical methods to solve the implicit Shockley equation or to find the maximum power point values of an experimental IV-curve. Thus, Muzzillo's FOM may be considered implicit to a large extent as well. We furthermore note that, when relying on experimental maximum power point data, Muzzillo's FOM should be the better choice as compared to our approximate FOM, as long as the sheet resistance of the TCE remains below the critical sheet resistance.^[7] In fact,

Muzzillo's FOM is—specifically with our modified version—very precise up to about the transition sheet resistance. As shown in Figure 1, the difference between the $\phi_{\text{Muzzillo,mod}}$ and ϕ_{Anand} up to $100 \Omega \text{ square}^{-1}$ (relevant to functional electrodes) is below 1%, and thus provides a remarkable agreement between the modified version of Muzzillo and ϕ_{Anand} . These FOMs thus mutually confirm each other within this region of sheet resistances. However, once sheet resistances exceed the critical sheet resistance,^[7] Muzzillo's FOM initially shows a large deviation and then delivers unphysical values below zero. It is the strength of the exact FOM to still provide physically meaningful values for solar cell performance, even at sheet resistances that are not meant for applications any longer.

This consideration brings us to the center or focal point of the discussion raised via Muzzillo's comment: as the same modifications and extensions of the exact FOM enable its successful application to a wide variety of application cases, what remains unresolved is then rather a philosophical question. While Muzzillo and others have invested large efforts into precisely describing the limiting effects of TCEs and grids for many individual scenarios, the exact FOM was introduced to put an end to arbitrary approximations by directly considering the photovoltaic impact of the TCE's material properties—optical transmittance and electrical conductance. From this perspective, Muzzillo's FOM is not too different from the exact FOM, as it considers the TCE's material properties within the framework of a linear perturbation theory.

In the case of the quoted 0.5 cm solar cell length the deviation between the network model and the one diode model for the exact FOM is very small ($\leq 1\%$) within the range of up to $100 \Omega \text{ square}^{-1}$ (see Seeland & Hoppe).^[11] At $100 \Omega \text{ square}^{-1}$, Muzzillo's FOM reaches a deviation of $\approx 5\%$ with regards to the exact FOM, thus rendering a deviation to the network model, including distributed series resistance considerations, of about 4% at minimum. However, modifying Muzzillo's FOM beyond linear effects brings it into close agreement with the exact FOM even up to $100 \Omega \text{ square}^{-1}$.

One can always find conditions for solar cells and solar modules, that require additional calculational effort to be correctly captured by the simulation; however, for most basic applications—specifically for those groups working on tiny solar cells for record efficiencies, the exact FOM provides an absolute number indicating the attainable photovoltaic performance as a fraction of the Shockley-Queisser Limit. In fact, the exact FOM moves one step further beyond the SQL, since solar cell length is getting considered. And if that requires to use of numerical methods and even solvers for implicit equations, we are at this stage of computerization and digitalization of the society ready to cope with that challenge.

In the end, we believe both approaches have a right to exist—if not coexist—and provide their own meaning and sense: caring about the details of specific application cases for very practical reasons, as well as caring about the broader picture of what ultimately governs the maximum performance of solar cells and modules that use TCEs. To reconcile both efforts and to support further development of TCEs for photovoltaics, we plan to offer the combined benefits to researchers all over

the world in the form of an online tool that will compute the exact FOM for their TCE and anticipate solar module geometry.

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Conflict of Interest

The authors declare no conflict of interest.

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