Singlet fission and tandem solar cells reduce thermal degradation and enhance lifespan

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Abstract
The economic value of a photovoltaic installation depends upon both its lifespan and power conversion efficiency. Progress toward the latter includes mechanisms to circumvent the Shockley-Queisser limit, such as tandem designs and multiple exciton generation (MEG). Here we explain how both silicon tandem and MEG-enhanced silicon cell architectures result in lower cell operating temperatures, increasing the device lifetime compared to standard c-Si cells. Also demonstrated are further advantages from MEG enhanced silicon cells: (i) the device architecture can completely circumvent the need for current-matching; and (ii) upon degradation, tetracene, a candidate singlet fission (a form of MEG) material, is transparent to the solar spectrum. The combination of (i) and (ii) mean that the primary silicon device will continue to operate with reasonable efficiency even if the singlet fission layer degrades. The lifespan advantages of singlet fission enhanced silicon cells, from a module perspective, are compared favorably alongside the highly regarded perovskite/silicon tandem and conventional c-Si modules.

KEYWORDS
module temperature, Perovskite tandem, singlet fission

1 | INTRODUCTION

At present, wafer-based silicon modules are the dominant PV technology. These modules are usually provided with warranted lifetimes of 25 years. Progress in all areas of PV module manufacturing will reduce costs, but there remains significant scope to reduce the cost of PV electricity through improved power conversion efficiency and by increasing the module lifespan beyond the present 25 years.

The power conversion efficiencies of conventional cells are approaching the single threshold Shockley-Queisser (SQ) efficiency limit. Efforts to circumvent this limit include the introduction of multiple absorbing thresholds, which can take several forms. Tandem cells incorporate a second junction with a larger bandgap material (such as perovskites or III-Vs), and excitonic methods exploit multiple-exciton generation processes, such as singlet fission. The process of singlet fission in molecular semiconductors is well established, whereby a photo-excited singlet exciton undergoes fission into two triplet excitons, producing twice the electronic charge carriers for each absorbed photon.

While both these approaches increase efficiency, little thought has been given to their impact on thermal load, particularly when considering the degradation present in realistic devices. Here, we...
provide a model for understanding the impact of multiple absorbing thresholds on thermal load. We investigate the thermal load reduction from both tandem and singlet fission cells, and estimate the resulting improvement to the lifespan of a module made from those cells.

2 | METHOD

A schematic of the heat generation and loss processes in a solar module is shown in Figure 1. Most of the incident sunlight is absorbed in the module, largely in the cells rather than the encapsulant. The encapsulation is an optically transparent glass coversheet with a thin absorbing rear polymeric coversheet placed against the cells. Accounting for optical reflection, the power that enters the module ($P_{in}$) is reduced compared to the incident sunlight. Heat is conducted to both module surfaces from where it is dissipated by free ($P_{c,free}$) and forced ($P_{c,forced}$) convection as well as radiatively ($P_{rad}$).

Prior to 2016, module thermal performance was characterized in terms of a single parameter, the nominal operating cell temperature (NOCT), defined as the temperature reached by open circuited cells in a module under a defined set of operating conditions: irradiance of 800 W·m⁻², 20°C ambient temperature, wind speed of 1 m·s⁻¹, and mount tilt angle of 45°. Following the introduction of an updated standard (IEC61215: 2016), NOCT has now been replaced by the NMOT (nominal module operating temperature), which is defined slightly differently. Consistent with the name change, the back-of-module temperature is now measured rather than the cell temperature. The standard reference environment remains unchanged with one major caveat, the electrical operating conditions are set to when the module load matches peak power generation instead of open-circuit. Experimental differences are small, with measurements taken at NREL (National Renewable Energy Laboratory, USA) over 38 days for a single module giving NMOT = 48.7 ± 1.7°C and NOCT = 47.9 ± 1.3°C.

In our thermal model for c-Si based devices, solar photons absorbed above the bandgap ($\lambda < 1200$ nm) convert their energy to both electricity and excess heat, while all photons absorbed below the bandgap contribute to heat only. Applying these assumptions to our thermal model, the thermalization losses within a solar module become $P_{in} - P_{elec}$. The solar module operating temperature was determined by solving $P_{in} - P_{elec} = P_{rad}(T) + P_{convec}(T)$ where these parameters are defined in Figure 1. The total incident irradiated solar power absorbed by a photovoltaic cell is given by:

$$P_{in} = \int_0^\infty \alpha(\lambda) \times b_{AM1.5G}(\lambda) d\lambda$$

where $b_{AM1.5G}$ is the solar spectral irradiance for sunlight under the standard air-mass 1.5 global condition, giving a total irradiance of 1,000 W·m⁻². For a c-Si solar photovoltaic module, the absorptivity $\alpha(\lambda)$ is well approximated by 95% across the entire solar spectrum and, accounting for the defined incident irradiance for NMOT (800 W·m⁻²), the total available solar power becomes

$$P_{in} = 0.95 \times 0.8 \int_0^\infty b_{AM1.5G}(\lambda) d\lambda$$

The outgoing power comprises contributions from electrical power generation, gray-body radiation, and convection, resulting in

![Solar module energy balance](image-url)
The electrical power delivered by the Si cell $P_{\text{elec,SI}}$ is given by

$$P_{\text{elec,SI}} = \int_0^{\lambda(E_g)} (1 - d_S)N_{in}(\lambda)V_{mp,SI} d\lambda$$

where $V_{mp,SI}$ is the voltage at the maximum power, estimated as 0.6 V, and $E_g$ is the silicon band gap. $(1 - d_S)N_{in}(\lambda)$ represents the photo generated current, $d_S$ accounts for power degradation of Si module, $N_{in}(\lambda)$ is the spectral photon flux, defined below:

$$N_{in}(\lambda) = \frac{0.95 \times 0.8 \frac{b_{\text{abs, sc}(\lambda)}}{hc/\lambda}}{2}$$

$h$ is the Planck constant, and $c$ is the speed of light.

In the case of general singlet fission layer degradation, all the photon generation above the singlet fission threshold is subjected to the singlet fission degradation rate. The optimum energetic configuration for a singlet fission solar cell is endothermic, owing to the increase in entropy following the fission process. The endothermic singlet fission process is accounted by $(1 - d_{SF})N_{in}(\lambda)\Delta E$. At energies below the singlet fission threshold, photon generation proceeds as for the conventional Si solar cell, giving

$$P_{\text{elec, SF}} = \int_0^{\lambda(E_{fiss})} 2 \times (1 - d_{SF})(1 - d_S)N_{in}(\lambda)V_{mp,SI} d\lambda + (1 - d_{SF})N_{in}(\lambda)\Delta E d\lambda + \int_{\lambda(E_{fiss})}^{\lambda(E_S)} (1 - d_S)N_{in}(\lambda)V_{mp,SI} d\lambda$$

where $d_{SF}$ accounts for the degradation of the singlet fission layer, and the optical energy threshold for singlet fission is $E_{fiss}$.

For the perovskite/Si tandem cell, independent degradation rates are used, and $d_{PSK}$ describes the degradation rate of perovskite layer. $V_{mp,PSK}$ is the perovskite maximum power voltage (0.8 V).

$$P_{\text{elec,PSK/SI}} = \int_0^{\lambda(E_{PSK})} (1 - d_{PSK})N_{in}(\lambda)V_{mp,PSK} d\lambda + \int_{\lambda(E_{PSK})}^{\lambda(E_S)} (1 - d_S)N_{in}(\lambda)V_{mp,SI} d\lambda$$

The net radiative power loss, $P_{rad}$, will depend on the orientation of the panel with respect to the sky and ground, and their respective temperatures:

$$P_{rad} = \sigma \left( \frac{1 + \cos^2 \beta}{2} \right) \epsilon_{\text{sky}} T_{\text{sky}}^4 + \frac{1 - \cos^2 \beta}{2} \epsilon_{\text{ground}} T_{\text{ground}}^4 - \epsilon_{\text{module}} T_{\text{module}}^4$$

where $\sigma$ is the Stefan–Boltzmann constant, $\beta$ is the angle of elevation for the module from the horizontal and taken to be 45°, $\epsilon_{\text{module}}, \epsilon_{\text{sky}},$ and $\epsilon_{\text{ground}}$ are emissivity values for solar module, sky, and ground, taken to be 0.84, 0.82, and 0.95, respectively. The sky temperature, $T_{\text{sky}}$, is calculated by:

$$T_{\text{sky}} = 0.0552 \times T_{\text{ambient}}$$

Where $T_{\text{ambient}}$ is the ambient temperature. Convective heat transfer per unit module area is expressed by

![Figure 2](image-url) Comparing conventional and tandem devices. (A) Solar spectral irradiance AM1.5G overlaid with the heat load spectra for various cell architectures. The inset shows the relevant 300 to 1200 nm wavelength range. (B) The carrier generation process in each configuration and the equivalent circuit. BC denotes a buck converter which halves the voltage and doubles the current, representative of the singlet fission process. [Colour figure can be viewed at wileyonlinelibrary.com]
where the heat transfer coefficient for forced convection at wind speeds of 1 m·s⁻¹ is assumed to be \( h_{c,\text{forced}} = 9.5 \, \text{W} \cdot \text{m}⁻² \cdot \text{K}⁻¹ \). The heat transfer coefficient for free convection from a vertical plane in air can be approximated by \( h_{c,\text{free}} = 1.31 \sqrt{T_{\text{module}} - T_{\text{ambient}}} \)

### 3 RESULTS AND DISCUSSION

#### 3.1 Heat load and lifespan

To validate the model, the working voltage for a hypothetical solar cell is estimated with \( V_{\text{mp}} = 0.6 \text{ V} \) for a c-Si solar cell. Solving the energy balance equation \( P_{\text{in}} = P_{\text{out}} \), and calculating \( P_{\text{in}} \) and \( P_{\text{out}} \) by Equation 2 and 3 respectively, results in \( T_{\text{module}} = 46.1 \, \text{°C} \), in line with the rule of thumb that modules operate 20–30°C above the ambient temperature.

We next apply the model to two well-studied approaches to multiple threshold devices: a perovskite on silicon tandem and a tetracene-based singlet fission device. For the perovskite/Si tandem cell, the working voltage is taken as 1.4 V (0.8 and 0.6 V for the perovskite and Si junctions, respectively). Singlet fission in the molecular semiconductor tetracene is well-known to generate two triplet excitons that are energetically matched to the silicon bandgap. The singlet fission energy threshold is assumed to be at 530 nm, slightly endothermic with \( \Delta E = 0.2 \, \text{eV} \).

The operating temperature of these two configurations are calculated as \( T_{\text{PSK/Si}} = 44.4 \, \text{°C} \) and \( T_{\text{SF/Si}} = 43.7 \, \text{°C} \), showing a reduction in temperature \( \Delta T = 1.7 \, \text{°C} \) and \( \Delta T = 2.4 \, \text{°C} \) compared with the conventional Si module, respectively. c-Si module lifetime is generally found to double for every 10°C reduction in temperature, such that the lifetime \( L \) at temperature \( T \) can be related to a nominal lifetime at \( T_0 \) by \( L = L_0 \frac{T}{T_0}^{2/3} \). This equates to an increase in lifetime of 3.1 years (12%) for the perovskite tandem and 4.5 years (18%) for the tetracene-based SF cell. A comparison of key parameters for the three structures discussed above are shown in Table 1.

#### 3.2 Degradation

Degradation is inevitable in all components of a PV module. Conventional silicon modules operating in the field typically suffer a 0.4% per annum degradation rate; commercial thin-film technologies (CdTe, CIGS) typically degrade roughly twice as quickly.

While it is difficult to realistically predict actual degradation rates for new top materials at this stage, perovskite materials are known to suffer from stability issues that require, at a minimum, very effective encapsulation; the same will also be true for tetracene-derived singlet-fission materials. A recent study on the efficiency premium offered by a perovskite silicon tandem established a 3.5% per annum degradation rate as the maximum acceptable value to achieve an overall energy yield advantage compared with conventional silicon cells.

| Structure          | Initial efficiency (%) | \( P_{\text{elec}} \) (W) | \( P_{\text{rad}} \) (W) | \( P_{\text{conv}} \) (W) | Temperature (°C) | Lifespan (years) |
|--------------------|------------------------|---------------------------|--------------------------|---------------------------|------------------|-----------------|
| c-Si module        | 26.5                   | 211.7                     | 198.7                    | 349.9                     | 46.1             | 25              |
| PSK/Si module      | 31.0                   | 248.1                     | 188.0                    | 324.3                     | 44.4             | 28.1            |
| SF/Si module       | 32.8                   | 262.3                     | 183.8                    | 314.2                     | 43.7             | 29.5            |
Degradation of singlet-fission layers is recognized as one of the primary obstacles to the development of a commercially viable device. Although the current matching considerations and the consequences of top cell degradation seen in two-terminal tandem series multi-junction devices are not present in the singlet fission device, intrinsic photodegradation can still play a significant role in the commercial viability. It should be noted that the perovskite/Si tandem cell considered here, as a three-terminal device, is not subject to current matching considerations. A two-terminal perovskite/Si tandem cell would have significantly more complicated degradation-related temperature properties as degradation of the top layer would reduce the overall current flow, leading to increased thermalization in the c-Si.

While considerable efforts are underway to identify stable alternatives, tetracene is currently the only singlet fission material to have demonstrated energy transfer into silicon. Importantly, ditetracene (the degradation product of tetracene under oxygen- and water-free conditions) is transparent to solar radiation. Therefore, although degradation occurs, a singlet fission/silicon solar cell will revert to the primary underlying silicon solar cell (in the ideal case where degradation does not enhance light scattering), having gained both efficiency and lifetime benefits for the stable duration of the tetracene layer. This compares favorably against two-terminal tandem series multi-junction devices where current matching considerations ensure that degradation of the top cell significantly impacts device performance.

Since the degradation product of tetracene singlet fission film is transparent to solar radiation, it will revert to standard c-Si solar cells after the “benign” degradation process, and the temperature and efficiency of tetracene singlet fission/Si module converges to that of the conventional c-Si module. In order to account for this benign generation we must change the first term of Equation 6 such that

\[ P_{\text{elec, SF}_\text{benign}, \text{Si}} = \left( 1 - d_{\text{Si}} \right) N_{\text{in}}(\lambda) V_{\text{mp}} + \left( 1 - d_{\text{SF}} \right) N_{\text{in}}(\lambda) \Delta E \]

In Figure 4 we show the effect of degradation on module temperature, efficiency and cumulative energy yield for conventional silicon, perovskite/silicon, and singlet fission/silicon solar modules with a lower bound for degradation at 0.5%/year and an upper bound of 3.5%/year for the perovskite and singlet fission components. At the beginning of life, all cells lie at the lowest temperature points, corresponding to the lower left-hand side of Figure 4A. As time

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

**Figure 4** The degradation effect of different device architectures. (A) Increase in module temperature, (B) decrease in module efficiency, (C) cumulative energy yield, and (D) cumulative energy yield above 20 years lifetime plotted for modules composed of conventional c-Si, singlet fission/silicon, and perovskite/silicon. The c-Si degradation rate is 0.4% per annum; singlet fission/silicon and perovskite/silicon degradation rates have a lower bound of 0.5% per annum and an upper bound of 3.5% per annum [Colour figure can be viewed at wileyonlinelibrary.com]
progresses, these cells will degrade at variable rates, but not exceeding the bounds of the shaded region to some end of life state. Similarly, all cells show highest efficiency at beginning of life in Figure 4B, and gradually degrade with time. Assuming five peak-Sun hours of irradiance in a day, the cumulative yearly energy yield of each devices is calculated and plotted in Figure 4C,D. Given the typical 0.4%(relative)/°C temperature coefficient of module efficiency, a 5–10°C reduction in module operating temperature corresponds to a 2%–4% gain in annual energy production. However, a much significant energy gain is the lifespan extension of the module. As discussed earlier in section “Heat Load and life span”, reducing the module operating temperature by 10°C doubles the module life and hence more than doubles energy yield. Therefore, we neglected the temperature effect on module efficiency in our model, considering it is significantly small compared with the lifespan effect. With a higher beginning of life efficiency, both perovskite/silicon and singlet fission/silicon solar modules show the advantage in lifetime energy yield with lower degradation rates. Especially the benign tetracene based singlet fission/silicon module always surpass the performance of a conventional silicon module since reverts to the baseline crystalline silicon module thermal profile after degradation.

The degradation path that a module may follow given the assumed limits to cell degradation stated above is illustrated in Figure 5A–C. The module temperature has been calculated for different degradation states for each of the respective perovskite/silicon tandem, generic singlet fission/silicon (where the degradation product is not transparent to solar radiation), and benign tetracene singlet fission/silicon (where the degradation product is transparent to solar radiation). For reference, c-Si solar modules typically suffer <20% cumulative degradation during a 25 year lifespan, while up to 100% degradation is modeled for perovskite and singlet fission materials in each configuration.

Within the degradation envelope considered in this study, the perovskite/silicon and singlet fission/silicon modules operate at lower temperatures for the first 5 years of operation. Thereafter modules where degradation takes place in the upper layer (either perovskite or singlet fission) the thermal benefit is marginal or serves to increase the overall module temperature. Elevated module temperatures will not only accelerate the degradation rate of the base silicon cell but will also increase the degradation in the perovskite or singlet-fission layer, leading to compounding temperature increases. This does not take place with the benign, tetracene based singlet fission layers.

![FIGURE 5. Module operating temperature resulting from cumulative degradation. (A) Perovskite/silicon, (B) general singlet fission/silicon modules, and (C) “benign” tetracene singlet fission/silicon. The shaded region corresponds to the degradation rate of 0.5%–3.5% per annum. The inset diagrams depict the carrier generation processes for beginning and end of life. [Colour figure can be viewed at wileyonlinelibrary.com]](image)
where degradation in the singlet fission layer reverts to the baseline crystalline silicon module thermal profile but with a few years of operation at lower temperature and hence lower silicon degradation rate.

4  |  CONCLUSIONS

The opportunity for multiple threshold silicon solar cells to increase the spectral efficiency compared with conventional silicon solar cells is well known. Here, we have shown that there are also ancillary benefits to the approach in terms of lower module temperature and resilience under degradation. At low degradation rates, both perovskite-based tandems and singlet-fission cells reduce thermal load and increase the lifetime of the primary silicon module. At larger degradation rates, tetracene-based singlet fission cells still outperform conventional cells as the benign nature of the degradation product is not detrimental to the performance of the primary silicon cell. In all these cases there exists the potential to significantly reduce the cost of energy produced by solar photovoltaic systems by both increasing efficiency and lifespan.

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CONFLICT OF INTEREST

There are no conflicts to declare.

AUTHOR CONTRIBUTIONS

The ideas were conceived by T.W.S., N.J.E-D and M.A.G. Singlet fission degradation data was supplied by M.J.Y.T., D.R.M. and A.J. B, perovskite data by Y.J. The calculations were performed by Y.J. and verified by T.W.S., N.J.E-D and M.A.G. The first draft of the paper was written by Y.J. with subsequent revision by M.J.Y.T, D.R.M and M.P.N. The figures were prepared by Y. J, N.J.E-D and M.J.Y.T. All authors discussed the results and commented on the manuscript.

DATA AVAILABILITY STATEMENT

The authors declare that the data supporting the findings of this study are available within the paper. Further data beyond the immediate results presented here are available from the corresponding authors upon reasonable request.

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