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Article

Improved Thermal Stability under High Power Conversion Efficiency Condition in Inverted Ternary Organic Solar Cells with Three Different Electron Transport Layers

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Abstract: The efficiency of organic solar cells (OSCs) is influenced by various factors, among which environmental temperature plays a significant role. Previous studies have shown that the thermal stability of these cells can be enhanced by incorporating a third component into their structure. Ternary organic solar cells, in particular, have shown promising results in improving thermal stability. A well-designed electron transport layer (ETL) can significantly bolster thermal stability by facilitating efficient charge transport and reducing charge recombination. In this study, we investigated the effect of temperature on the efficiency of inverted ternary structures, ranging from 300 K to 400 K. The structures examined include FTO/SnO₂/PM6:D18:L8-BO/PEDOT: PSS/Ag, FTO/Spiro-OMeTAD/PM6:D18:L8-BO/PEDOT: PSS/Ag, and FTO/PC60BM/PM6:D18:L8-BO/PEDOT: PSS/Ag. Through simulations employing three different electron transport layers—SnO₂, Spiro-OMeTAD, and PC60BM—we observed that at 340 K (65°C) these structures maintained 92% of their original high efficiency of ~20% at 300 K, which represents a high level of thermal stability under the high PCE condition.

Keywords: organic solar cells; ternary structures; operating temperature; electron transport layer; power conversion efficiency

1. Introduction

Nowadays, the international community faces a multifaceted global energy challenge, closely intertwined with the issue of global warming primarily induced by greenhouse gases, especially CO₂, in the atmosphere [1–6]. Fossil fuels, as the principal sources of approximately 90% of CO₂ emissions and over 70% of all greenhouse gas emissions worldwide, are significant drivers of climate change [7–12]. These gases trap solar heat, contributing to a rise in global temperatures [13–16].

Addressing global warming necessitates sustainable interventions, including reducing air pollution, which can be achieved through adopting renewable energy sources, minimizing waste, and conserving natural resources [13,17–19]. In this context, photovoltaic energy emerges as a sustainable and long-lasting solution that could mitigate the impacts of global warming [20–22]. OSCs, in particular, represent a promising alternative to traditional silicon-based panels due to their environmental benefits and cost-efficiency in production [23]. Their widespread adoption could significantly enhance global solar energy utilization and reduce the ecological impacts of human activities [20]. Particularly, the role of OSCs in reducing global warming effects is increasingly recognized [23–25]. Although their potential benefits are substantial, further research and development are critical to improve their efficiency and thermal stability [26–30].

Improving the thermal stability of OSCs is vital for advancing their commercial viability and operational reliability. Ongoing research focuses on developing new materials and structures that optimize both power conversion efficiency (PCE) and thermal stability. Recent research by Zhang et

al.[29] highlights the efficacy of the ternary blend approach in enhancing the thermal stability of OSCs, and it was found that the PCE of the device at 75°C remained on average 80% of its PCE at 25°C. Key contributors to the charge-transfer state energy in these cells were identified as PBDB-T and IDT-PDOT-C6, with ITC6-2F playing a crucial role in facilitating charge carrier transfers to IDT-PDOT-C6. This mechanism promotes the generation of additional excitons reaching the donor/acceptor interface, thereby achieving high-efficiency photoelectric conversion [29]. Anass et al.[31] studied the temperature influence on the performance of the solar cells with [(Cbz-Mth)-B-T]2-PCBM as an active blend, and they found that their structure kept 89% of its room-temperature PCE at 75°C. Another study done by Muhammad et al.[32] performed a simulation by stepping up the temperature from 300 K to 400 K to study the impact of the raised temperature on the efficiency of their solar cells with PBDB-T: ITIC-OE as a photoactive layer, and they obtained a loss ratio of the efficiency between the temperature range of 300-350 K to approximately 9%, which is a highly stable device. Moreover, the use of charge transport layers is paramount in improving the PCE of OSCs. These layers are essential for the efficient extraction and mobility of charges, while also preventing electron leakage, and are utilized extensively across various materials in the inverted OSCs architecture [33–37]. The previous studies found that the best thermal stability achieved by OSCs at 350 K is 9% in the studies done by Anass et al.[31] and Muhammad et al. [32], but PCEs achieved in these studies are 7.4% and 6.2%, respectively, which are relatively low. It is challenging to achieve high thermal stability and high PCE at the same time.

This study aims to explore how temperature variations affect the efficiency of inverted bulk heterojunction (BHJ) OSCs. Through simulations using three different electron transport layers—SnO₂, Spiro-OMeTAD, and PC60BM, we seek to derive insights into the performance and thermal stability of these cells, specifically focusing on analyses of their high thermal stability (quantified by the PCE loss ratio) at 350K under the high PCE of ~ 20%. This could potentially lead to the development of more efficient and stable OSCs in laboratory settings.

2. The Simulation Model

The simulation employs Oghma-Nano software, which utilizes a 1-dimensional drift-diffusion model for its electrical modeling. The model includes the solution of bi-polar drift-diffusion, charge carrier continuity, Poisson equation, and Maxwell-Boltzmann equation for free charge carrier statistics. The mathematical model can be found in the software's documentation for more detailed information [38]. The simulated structure of the device is FTO/SnO₂/PM6:D18:L8-BO/ PEDOT: PSS/Ag, and the input parameters and thickness are included in Tables 1–4.

Bi-polar drift-diffusion equations at the position for both electrons and holes are represented in Equations (1) and (2),

$$J_n = q\mu_e n_f \frac{\partial \phi}{\partial x} + qD_n \frac{\partial n}{\partial x} \quad (1)$$

and

$$J_p = q\mu_h p_f \frac{\partial \phi}{\partial x} + qD_p \frac{\partial p}{\partial x} \quad (2)$$

where $J_{n,p}$ is the electron and hole current density, q is the elementary charge, μ_e and μ_h are the mobilities of electrons and holes, respectively; D_n and D_p are the electron and hole diffusion coefficients, respectively; n is the density of electrons, and p is the density of holes. The charge carrier continuity equations are mentioned in Equations (3) and (4),

$$\frac{\partial J_n}{\partial x} = q(R_n - G + \frac{\partial n}{\partial t}) \quad (3)$$

and

$$\frac{\partial J_p}{\partial x} = -q(R_p - G + \frac{\partial p}{\partial t}) \quad (4)$$

where R_n and p are the recombination rate of electrons and holes, respectively; G is the generation rate.

The solution to Poisson's equation is used to determine the distribution of potential within the device, and it is represented as follows in Equation (5),

$$\frac{\partial \epsilon_0 \epsilon_r}{\partial x} \frac{\partial \varphi}{\partial x} = q(n - p) \quad (5)$$

where ϵ_0 , ϵ_r the constants of permittivity in free space and the relative permittivity are constant, respectively; φ is the voltage profile. The model applies Maxwell-Boltzmann statistics to solve free carriers statistics as mentioned in Equations (6) and (7),

$$n = N_c \exp \left(\frac{F_n - E_c}{K_B T} \right) \quad (6)$$

and

$$p = N_v \exp \left(\frac{E_v - F_p}{K_B T} \right) \quad (7)$$

where N_c , N_v are the constants of the effective density of states in the conduction and valence band of a semiconductor, $F_{n,p}$ are constants of the energy level of the Fermi level in the valence and conduction band of a semiconductor, E_c is the conduction band, E_v is the valence band, K_B is Boltzmann constant, T is the temperature.

The boundary conditions between the layer interfaces are represented as tunneling of electrons and holes through layer interfaces provided by Equations (8) and (9),

$$J_n = -qT_e [(n_1 - n_1^{eq}) - (n_0 - n_0^{eq})] \quad (8)$$

and

$$J_p = -qT_h [(p_1 - p_1^{eq}) - (p_0 - p_0^{eq})] \quad (9)$$

where T_e and T_h is the rate constants of tunneling of electrons and holes, respectively, $n_{1,2}$ is the number of electrons in the layers before and after the interface; $p_{1,2}$ is the number of holes before and after the interface; $n_{1,2}^{eq}$ is the equilibrium number of electrons in before and after the interface; $p_{1,2}^{eq}$ is the equilibrium number of holes in the layers before and after the interface.

More details of the above electrical model of our simulation can be found in Refs. [39–41].

3. Results and discussion

The density of current-voltage (J - V) was simulated under AM 1.5 G illumination with an intensity of 100 mW cm⁻² and a temperature range between 300 K and 400 K. The 1st structure is represented in Figure 1, and its results are presented in Figures 2 and 3. S1 showed the best PCE of 20.08 % at 300 K with a short circuit current (J_{sc}) of 27.4 mA cm⁻², an open circuit voltage (V_{oc}) of 0.89 V, and a fill factor (FF) of 82.2%, while the performance relatively decreased between the range of 310 and 400 K to reach a PCE of 15.53%, a J_{sc} of 27.35 mAcm⁻², a V_{oc} of 0.73 V, and a FF of 77.7% at 400 K. As the results mentioned, the major parameters that cause a reduction in the efficiency of OSCs during the enhancement of temperature are the V_{oc} and FF (see Figure 3 (b),(c)).

$$qV_{oc} = E_g - \Delta E \quad (10)$$

and

$$\Delta E = 2(E_{F,h} - E_{HOMO}^D) - K_B T \ln \left(\frac{\mu_e}{\mu_h} \right) \quad (11)$$

where $E_{F,h}$ is the energy corresponding fermi-level; E_{HOMO}^D is the electron donor energy.

The increase in temperature causes an augmentation of the energy loss (ΔE) as demonstrated by eq (11), and then the V_{oc} decreases as a result of that augmentation. The FF showed a negative correlation with temperature which decreased when the temperature increased from 300K to 400K (from 26.85°C to 126.85 °C). The performance decrease of FF due to the increase of the exponential of the temperature as demonstrated in eq (12). The J_{sc} showed relative stability under the increase in temperature. The reduction in J_{sc} can be attributed to the bandgap energy (E_g) effect.

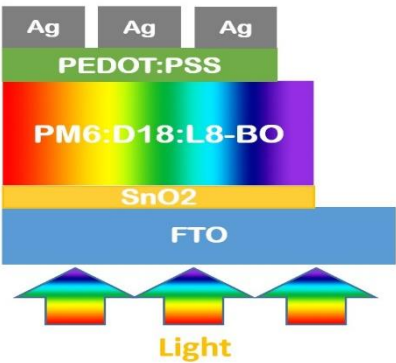


Figure 1. Schematic of the structure of an S1 OSC model under light exposure.

Table 1. Simulation parameters of Oghma-nano software.

Parameters	Values
Electron mobility (μ_e)	1.49e-07 m ² V ⁻¹ s ⁻¹
Hole mobility (μ_h)	1.42e-07 m ² V ⁻¹ s ⁻¹
Effective density of free electron (N_c at 300K)	1e26 m ⁻³
Effective density of free hole (N_v at 300K)	1e26 m ⁻³
n to P Recombination rate constant	1.15e-17 m ³ s ⁻¹
Free carrier statistics	Maxwell Boltzmann-numerical + analysis
Density of states distribution (DoS)	Complex
Free electron (n) to Trapped electron ($strap$)	1e-15 m ⁻²
Trapped electron (n_{trap}) to Free hole (P)	1e-20 m ⁻²
Trapped Hole (P_{trap}) to Free electron (n)	1e-20 m ⁻²
Free hole (P) to Trapped hole (P_{trap})	1e-15 m ⁻²
Number of traps (N_t)	5 Trap
Energy bandgap (E_g)	1.29 eV
Relative permittivity (ϵ_r)	3.0 a.u

Table 2. The thickness of the layers the blends S1 devices for optimum efficiency.

Layer	Thickness (nm)	Materials	Type
ITO	130	Oxides	Contact
SnO ₂	5	Polymers	Other
PM6:D18:L8-BO	80	Blends	Active
PEDOT: PSS	20	Oxides	Other
Ag	100	Metal	Contact

The FF determines the maximum power output by the OSC, it is defined below in eq (12),

$$FF = \frac{P_{max}}{V_{oc}J_{sc}} = \frac{V_m}{V_{oc}} \left[1 - e^{\frac{q}{AKT}(V_m - V_{oc})} \right] \quad (12)$$

where P_{max} is the maximum power delivered by the OSC, V_m is the maximum voltage, A is the ideality factor of the semiconductor.

As temperature rises, the J_{sc} experiences a marginal increase due to the reduction in E_g . Consequently, a greater number of photons possess adequate energy to generate electron-hole pairs. Nevertheless, the impact of this phenomenon is quite minor. The exponential relationship between temperature and the reverse saturation current of photovoltaic cells is observed in eq (13). Additionally, this factor can have an impact on the J_{sc} .

$$J(V) = J_0 \left[\exp \left(\frac{eV}{AKT} \right) - 1 \right] + J_{ph} \quad (13)$$

where J_0 is the reverse saturation current density, e is the charge of an electron, and J_{ph} is the photo-current density.

Table 3. The thickness of the layers blends S2 devices for optimum efficiency.

Layer	Thickness (nm)	Materials	Type
ITO	130	Oxides	Contact
Spiro-OMeTAD	5	Polymers	Other
PM6:D18:L8-BO	80	Blends	Active
PEDOT: PSS	20	Oxides	Other
Ag	100	Metal	Contact

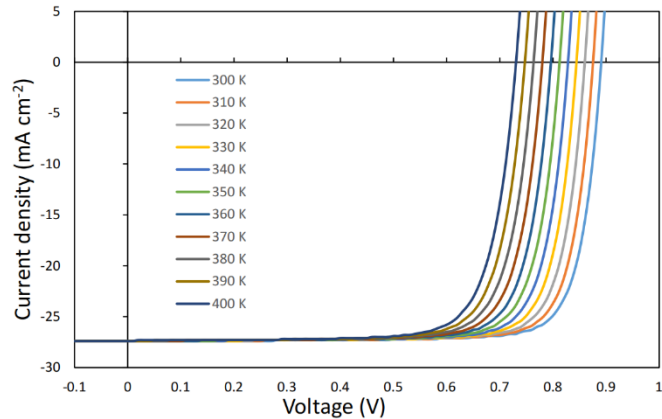


Figure 2. J - V the curves of the S1 device in the temeprature range of 300-400 K for a device area of 4.84 mm².

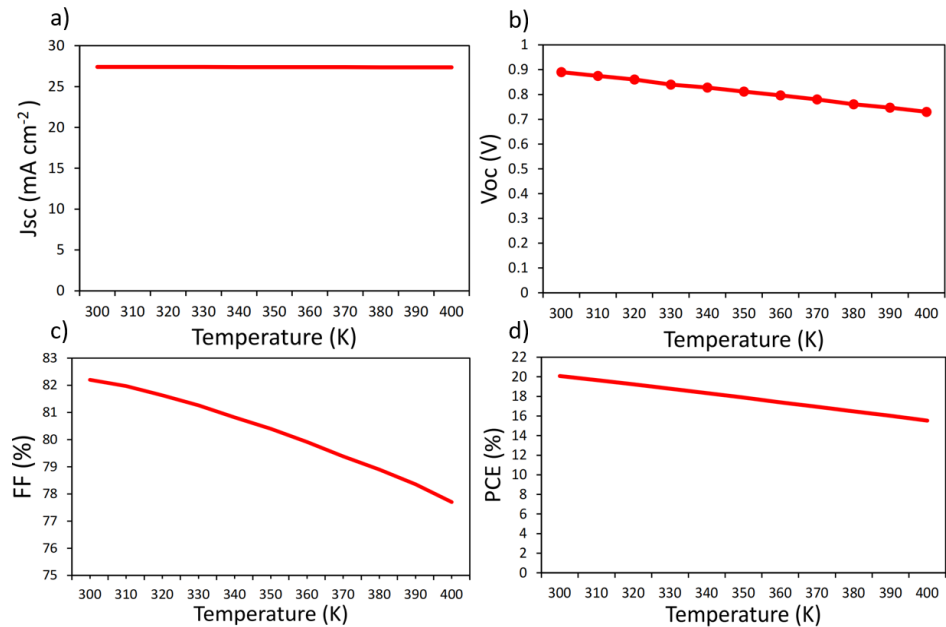


Figure 3. The temperature affects the performance of the S1 device: (a) J_{sc} as a function of temperature; (b) V_{oc} as a function of temperature; (c) FF as a function of temperature; and (d) PCE as a function of temperature.

The 2nd structure is presented in Figure 4, and its results are presented in **Figures 5 and 6**. S2 showed the best PCE of 20.11% at 300 K with a J_{sc} of 27.43 mA cm⁻², a V_{oc} of 0.89 V, and an FF of 82.3%, while the efficiency relatively decreased between the range of 310 and 400 K to reach a PCE of 15.55%, a J_{sc} of 27.38 mA cm⁻², a V_{oc} of 0.73, and a FF of 78% at 400 k. The results are slightly the same as S1.

As S2 results depicted, the major parameters that cause a decrease in the performance of this device during the enhancement of temperature are fundamentally the same as before V_{oc} and FF (see **Figure 6 (b), (c)**).

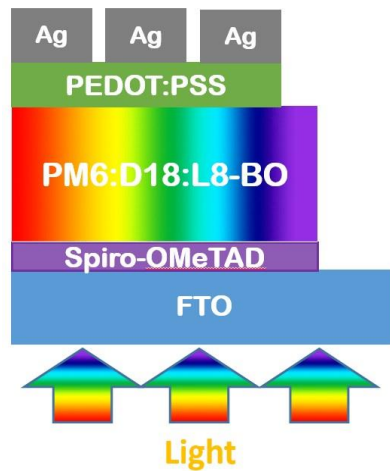


Figure 4. Schematic of the structure of an S2 OSC model under light exposure.

The increase in temperature caused an enhancement of the ΔE as demonstrated by eq (11), and then the V_{oc} decreased as a consequence of that enhancement. The diminution in J_{sc} can be attributed to the concept of E_g . As temperature rises, the short-circuit current, J_{sc} , experiences a slight increase due to the reduction in E_g . This decrease in bandgap energy enables a greater number of photons to possess sufficient energy for the generation of excitons. The reverse saturation current of photovoltaic cells exhibits an exponential growth pattern about temperature. Additionally, this factor can have an impact on the magnitude of the J_{sc} as demonstrated in eq (15).

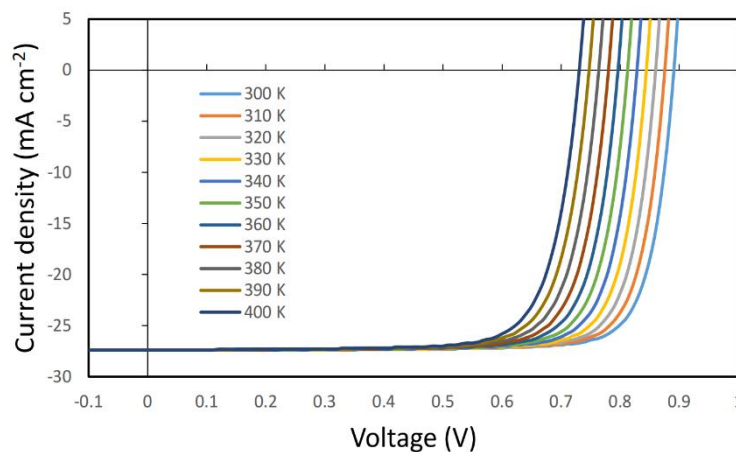


Figure 5. J - V curves of the S2 device in the temperature range of 300-400 K for a device area of 4.84 mm².

The 3rd structure is presented in Figure 7, and its outcomes are depicted in Figures 8 and 9. At a temperature of 300 K, S3 exhibited the highest PCE of 18.9%. This was accompanied by a J_{sc} of 25.8 mA cm⁻², a V_{oc} of 0.89 V, and a FF of 82.36%. However, when the temperature increased within the range of 310 to 400 K, the performance of S3 declined.

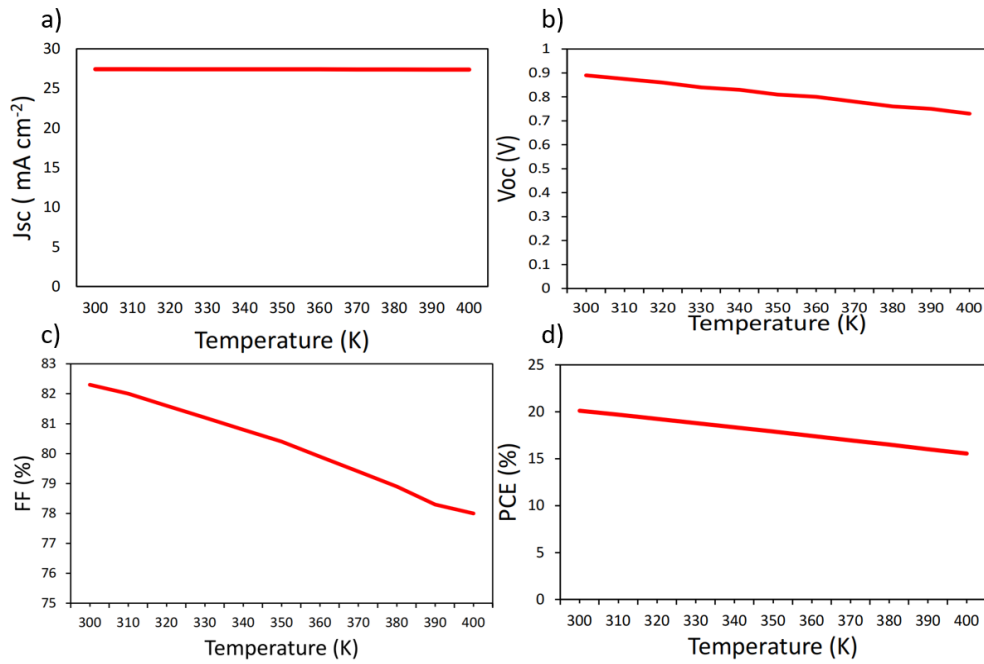


Figure 6. The temperature affects the performance of the S2 device: (a) J_{sc} as a function of temperature; (b) V_{oc} as a function of temperature; (c) FF as a function of the temperature; and (d) PCE as a function of temperature.

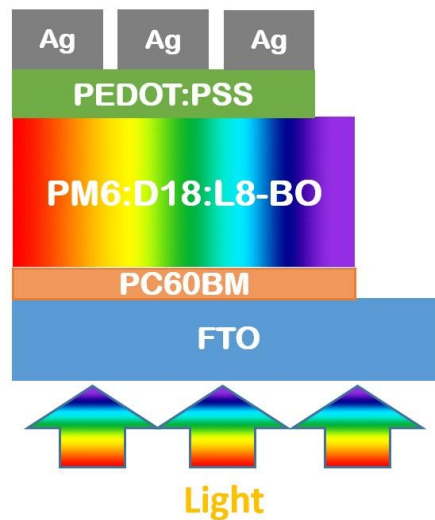


Figure 7. Schematic of the structure of an S3 OSC model under light exposure.

At 400 K, the PCE reduced to 14.6%, with a J_{sc} of 25.76 mA cm⁻², a V_{oc} of 0.73 V, and an FF of 78%. According to the findings from the S3 results, it can be observed that the primary factors contributing to the decline in device performance as temperature increases remain consistent with the V_{oc} , and FF (refer to **Figure 9** (b),(c)).

The rise in temperature leads to an increase in ΔE , as indicated by eqs (10, 11). Consequently, the V_{oc} decreases as a result of this increase. The decrease in J_{sc} can be ascribed to E_g . The J_{sc} undergoes a marginal augmentation as the temperature increases, primarily as a result of the decrease in the E_g . The reduction in bandgap energy facilitates an increased likelihood of photons possessing the necessary energy to produce electron-hole pairs. The temperature-dependent behavior of photovoltaic cells' reverse saturation current is characterized by an exponential increase as noted before in S1 and S2. Moreover, this particular element can influence the amount of J_{sc} , as exemplified in eq (11).

Table 4. The thickness of the layers of the blend S3 devices for optimum efficiency.

Layer	Thickness (nm)	Materials	Type
ITO	130	Oxides	Contact
PC ₆₀ BM	5	Polymers	Other
PM6:D18:L8-BO	80	Blends	Active
PEDOT: PSS	20	Oxides	Other
Ag	100	Metal	Contact

We observe that All the structures S1, S2, and S3 showed good performance under the temperature range between 300 and 340K (from 26.85°C to 66.85 °C) (see **Figure 5**), which they lost from their initial efficiency of only 8.8%, 8.8%, 8.9%, respectively. The performance parameters J_{sc} , V_{oc} , and FF showed slightly the same progression, while S3 depicted less PCE of 18.9% in comparison to S1 and S2 depicted 20.08%, and 20.11%, respectively. The inverted structure employing SnO₂ and Spiro-OMeTAD achieved the best PCE of 20.08% and 20.11%, respectively, in the other hand, the inverted structure using PC₆₀BM as an electron transport layer (ETL) showed less PCE . But, all devices worked efficiently in terms of performance.

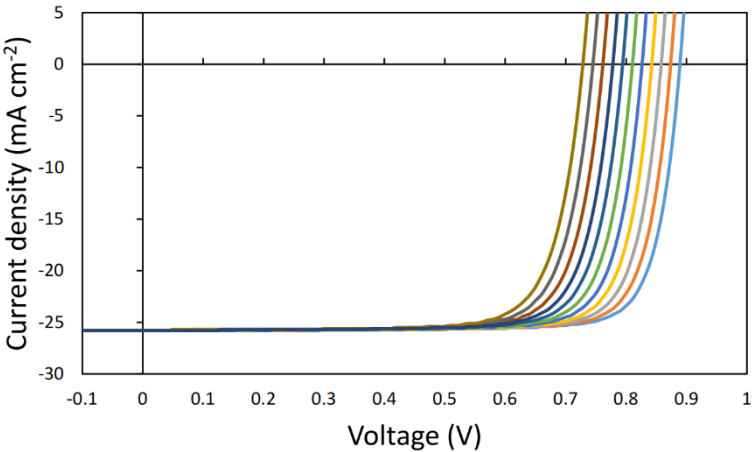


Figure 8. J - V curves of S3 device in the temperature range of 300-400 K for a device area of 4.84 mm².

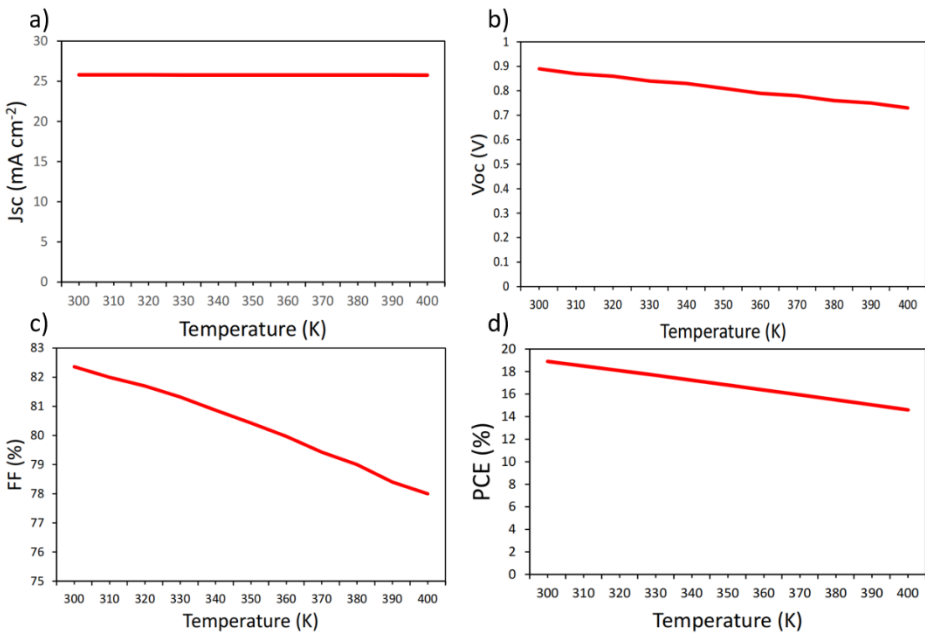


Figure 9. The temperature affects the performance of the S3 device: (a) J_{sc} as a function of temperature; (b) V_{oc} as a function of temperature; (c) FF as a function of the temperature; and (d) PCE as a function of temperature.

All the studies showed a stable loss ratio of 11 % between 300 k and 350 K according to the results mentioned in **Table 5**. We investigated our results of the loss ratio of PCE in the function of raising the temperature with some other numerical studies as mentioned in **Table 6**. The current study demonstrated the thermal stability of a device with a loss ratio of 11% throughout a temperature from 300 K to 350 K, in contrast to the results of Khan et al. and Kim et al., whose studies reported loss ratios of 15% and 17%, respectively. The studies done by Anass et al. and Muhammad et al. have demonstrated an enhancement in thermal stability when compared to previous research. Furthermore, in addition to its thermal stability, our structure has demonstrated a much higher PCE than Anass et al. And Muhammad et al. Our structures can maintain high thermal stability at high PCE conditions, representing a notable advancement in the OSCs. Our structures can maintain high thermal stability at high PCE conditions of ~20%.

Table 5. The photovoltaic parameters of three devices (S1, S2, and S3) under different temperatures.

	$T(K)$	$T(^{\circ}C)$	$J_{sc}(mA\ cm^{-2})$	$V_{oc}(V)$	$FF(\%)$	$PCE(\%)$
S1	300	26.85	27.4	0.89	82.2	20.08
	310	36.85	27.4	0.875	81.97	19.66
	320	46.85	27.4	0.86	81.63	19.23
	330	56.85	27.4	0.84	81.26	18.78
	340	66.85	27.38	0.828	80.82	18.33
	350	76.85	27.38	0.812	80.4	17.88
	360	86.85	27.37	0.796	79.91	17.4
	370	96.85	27.37	0.78	79.38	16.95
	380	106.85	27.36	0.76	78.9	16.48
	390	116.85	27.36	0.747	78.35	16.01
	400	126.85	27.35	0.73	77.7	15.53
S2	300	26.85	27.43	0.89	82.3	20.11
	310	36.85	27.43	0.875	82	19.68
	320	46.85	27.42	0.86	81.6	19.25
	330	56.85	27.42	0.84	81.2	18.8
	340	66.85	27.41	0.83	80.8	18.35
	350	76.85	27.41	0.81	80.4	17.9
	360	86.85	27.41	0.8	79.9	17.43
	370	96.85	27.4	0.78	79.4	16.96
	380	106.85	27.4	0.76	78.9	16.5
	390	116.85	27.38	0.75	78.3	16
	400	126.85	27.38	0.73	78	15.55
S3	300	26.85	25.8	0.89	82.36	18.9
	310	36.85	25.8	0.87	82	18.5
	320	46.85	25.8	0.86	81.7	18.08
	330	56.85	25.79	0.84	81.32	17.67
	340	66.85	25.79	0.83	80.87	17.23
	350	76.85	25.78	0.81	80.43	16.81
	360	86.85	25.78	0.79	79.97	16.37
	370	96.85	25.77	0.78	79.43	15.93
	380	106.85	25.77	0.76	79	15.5
	390	116.85	25.77	0.75	78.4	15.05
	400	126.85	25.76	0.73	78	14.6

Table 6. Results of the *PCE* loss ratio (%) due to the temperature increment.

	<i>PCE</i> at 300K	<i>PCE</i> at 350K	Loss ratio %
Khan et al.[43]	5.75%	4.88%	15%
Anass et al.[31]	8%	7.4%	9%
Muhammad et al.[32]	6.82%	6.2%	9%
Kim et al. [25]	3.25%	2.7%	17%
Our study	20.11%	17.9%	11%

4. Conclusions

Our research demonstrated that the efficiency of inverted ternary organic solar cells incorporating SnO₂, Spiro-OMeTAD, and PC60BM is robust to temperature increases, sustaining less than 10% efficiency loss at 65°C compared to their baseline efficiency at 25°C. This finding underscores the commendable thermal stability of these structures. In addition, we achieved such thermal stability at the high *PCE* condition, indicating superb environmental adaptability of investigated structures. However, the configuration using PC60BM as the electron transport layer exhibits relatively lower *PCE*, highlighting an area for potential improvement. Future studies should expand on these preliminary findings with comprehensive evaluations focusing on the effects of operational temperature on newly synthesized organic semi-conductor materials to discover more stable structures and futher increase the thermal stability of the organic solar cells. This includes investigating charge mobilities, optimizing cooling systems, assessing material quality, and testing endurance under varying ambient temperatures.

Author Contributions: M.E.A.B.: conceptualization, methodology, writing original draft preparation, formal analysis, investigation and data curation; Q.W.: resources, writing review and editing; C.Z.: formal analysis, investigation, project administration, resources, funding acquisition and supervision. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement: The data are available upon reasonable requests from the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

q	The electrical charge
n	The concentration of free electrons
n_0	The equilibrium density of electron
$N_{c,v}$	The effective density of states in the conduction band
p_0	Equilibrium density of holes
P	Concentration of free holes
$J_{n,p}$	The current density of the electrons and holes
v_{th}	The thermal emission velocity of the carriers
$E_{c,v}$	The conduction and valence band
$E_{F,h,e}$	The energy corresponding fermi-level
$F_{n,p}$	The Fermi level in the conduction and valence band
$D_{n,p}$	The diffusion coefficient of the electrons and holes
R_n	The recombination rate of electrons
R_p	The recombination rate of holes
G	The carrier generation rate
K_B	Boltzmann constant
T	The Temperature
Greek Symbols	
ϵ_0	The permittivity of free space
ϵ_r	Relative permittivity

φ	The voltage profile
$\mu_{e,h}$	Electron and hole mobility
ΔE	The energy offset
ω	The angular frequency of the wave
λ	The wavelength
Superscripts	
D	Electron donor
A	Electron Acceptor

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