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Thin-Film Tandem Organic Solar Cells With Improved Efficiency

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ABSTRACT Seeking high power conversion efficiency is always a striving pathway for many researchers to commercialize organic solar cells (OSCs). Thus, it is essential to achieve high conversion efficiency to make solar oriented devices as truly green energy resource. In this paper, we report novel tandem structures based on organic materials in which homo and hybrid schemes are utilized. The homo tandem structures involve the well-known organic materials such as P3HT: PCBM and PTB7: PCBM as top and bottom absorber layers, while the hybrid tandem structure is made by the combination of P3HT: PCBM and PTB7: PCBM, respectively. Due to different absorption coefficients, simulation results indicate that the hybrid tandem structure exhibit large values for current density $J_{sc} = 22.60 mA/cm^2$, open circuit voltage $V_{oc} = 0.85V$, fill factor $FF = 68.21\%$, and efficiency $\eta = 13.96\%$, compared to homo tandem cell and proves to be more efficient and highly stable towards high temperatures, which indicates that the proposed structure is more suitable for practical applications.

INDEX TERMS Organic solar cell, tandem, homo, hybrid, efficiency.

I. INTRODUCTION

Organic solar cells (OSCs), also known as polymer solar cells, is of worth attention for many researchers due to their potential as an alternative to the existing commonly used inorganic solar cells. However, many potential applications of OSCs are still under restriction due to their low efficiency and low stability when compared to other kind of solar cells especially with inorganic solar cells. Therefore, for improving the efficiency of OSCs, great efforts have been devoted. In order to achieve this goal, different methods including device structure tuning [1], characterization [2], active materials modification [3] and annealing [4], [5], were firstly employed and then explored.

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In organic photovoltaics (OPV), large amount of sunlight energy is lost when single photoactive material is employed, resulting in poor efficiency. The loss of photons may be due to smaller photon energy compared to the bandgap of active material, which generally transmit through the cell without being absorbed. To counter these issues, tandem structure plays an important key role. In tandem structure multi-stacking layer structure is used which compose of different absorber layers either connected in series or in parallel [6]. The series connected multilayer structure provide an efficient mode to extract high value of η and V_{oc} [7]. The different absorber layers with different absorption coefficient are responsible for the absorption of photons with different energies levels. Various polymers and molecules combined together to form bulk material to make strong and broad spectral coverage [8]–[12]. So, tandem structures configured with multi absorber layers provide enhance absorption of photons. However, the tandem structure based on organic material has not been studied intensively due to complexity, so there is a great room to improve the performance of such tandem based organic devices.

D. W. Zhao *et al.*, demonstrated tandem OSCs and extracted the efficiency of 2.82% and 3.88% respectively [13]. The extracted efficiency is quite low in their cell, which cannot be utilized for practical applications because such low efficiencies cannot be considered as efficient structure. M. Riede *et al.*, reported the conversion efficiency of 6.07% [14]. However, there is still a room of improvement in their structure by utilizing different absorber layer, which shows high absorption and give higher conversion efficiency. J. Gilot *et al.*, fabricated and analyzed polymer tandem solar cell and extracted the conversion efficiency of 4.9% under AM 1.5 G [12]. Nevertheless, their extracted conversion efficiency is very small and can be improved more by utilizing different layers, which shows high absorption and transmission spectra. Z. Jiang *et al.*, fabricated and analyzed tandem structure, and reported a conversion efficiency of 1.6% and 3.4% for hetero and homo tandem structure respectively [7]. From the survey, we concluded that low efficiency is currently the main challenge in organic solar cells, which need to be countered for the advancement in organic photovoltaic technology.

In this paper, we investigated three cases for tandem structures and compared their performance parameters. The investigated tandem structures are (i) ITO/Zno/P3HT:PCBM/P3HT:PCBM/PEDOT:PSS/ZnO/P3HT:PCBM/MoO_3/Al (ii) ITO/ZnO/PTB7:PCBM/PEDOT:PSS/ZnO/PTB7:PCBM/MoO_3/Al (iii) ITO/PEDOT:PSS/P3HT:PCBM/TiO_2/PEDOT:PSS/PTB7:PCBM/TiO_2/Al. The case (iii) structure exhibits the high conversion efficiency as compared to case (i) and (ii) due to different absorber materials.

II. DEVICE MODELING

The structure of the proposed tandem solar cell is shown in Fig. 1, which consists of nine different effective layers. P3HT: PCBM and PTB7: PCBM are used as photoactive absorber layers because they have the ability to absorb enormous amount of incident photons even in low light conditions, good thermal stability and low cost manufacturing processing [15]. The thickness of upper and lower sub-cells are optimized to obtain high performance parameters for the proposed tandem OSCs structures. For top and bottom electrodes, indium tin oxide (ITO) and aluminum (Al) are used. The ITO has dual property in nature i.e., allow incident light to the photoactive layer in the visible spectrum range due to high work function and also collect carrier when used as an electrode [16]. On the other hand, Al has high reflectance at room temperature, low manufacturing cost, excellent thermal stability, low specific resistivity, acceptable adherence and uniformity along flat substrate [17]. Zinc oxide (ZnO) and titanium dioxide (TiO_2) materials, which have low resistivity, are used as window layer and electron transport layer (ETL)

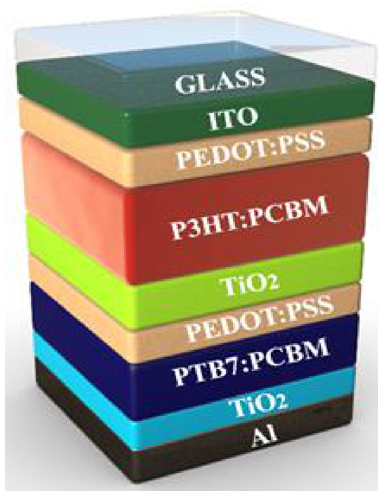

FIGURE 1. Schematic of hybrid tandem OSC structure.

for better light transmission, high electron mobility and also act as hole blocking layer (HBL) to reduce the recombination losses. Furthermore, they have excellent optical activity, non-toxicity, high stability, low water angle contact, charge transport and good optoelectronics property [18]–[22]. Different buffer layers including PEDOT:PSS and Molybdenum trioxide (MoO_3) are used as hole transport layer (HTL), as they exhibits better electrical conductivity, good stability in high temperatures, excellent hole transporting ability as well as low cost and high mechanical flexibility [23]–[26]. Addition to this, at the interference the buffer layer helps to reduce the recombination [21].

All simulations are carried out in the environment of one dimensional (1-D) general purpose photovoltaic device model (GPVDM) simulation tool [27], which is an open source tool for PV research community. The advantage of this simulation tool is that, it has the ability to calculate electrical and optical properties of OSCs more precisely and accurately as compared to others [28]–[30]. In this study, we have worked on the electrical model of the proposed OSCs device.

The short circuit current density (J_{sc}) of tandem based cell is given by photocurrent densities produced by the cell junctions. The analytical model based carrier continuity equation is considered by utilizing electric field dependent escape probability and transport carrier through photo absorber active layer. The photocurrent density in response to incoming solar radiations can be written as [31]–[33]

$$J_{phl}(V) = \int_0^{\infty} \left\{ J_p(\lambda, V) + J_n(\lambda, V) \right\} d\lambda \quad (1)$$

where, the density of photocurrent for electrons and holes is given by $J_n(\lambda, V)$ and $J_p(\lambda, V)$

For electrons, the steady-state continuity equation can be describe as

$$\frac{\partial}{\partial t}(\delta_n) = \mu_n F \frac{\partial}{\partial x}(\partial_n) + D_n \frac{\partial^2 \delta_n}{\partial x} + Ge^{-X\alpha(\lambda)} - \frac{\partial_n}{\tau_n} = 0 \quad (2)$$

where, photo generated electron concentration is represented by δ_n, the distance from the front contact along the absorber layer is symbolize as x, the electron mobility is represented as μ_n, the electron diffusion is represented as D_n and F is the electric field.

For electrons, the photocurrent density is given by

$$J_n(\lambda, V) = \frac{e}{W}\left\{\mu_n F \int_0^W \delta_n dx - Dn \int_0^W \frac{\partial \delta_n}{\partial x} dx\right\} \quad (3)$$

where, n in the subscript denotes electrons.

Similarly, for holes, the photocurrent densities is given by

$$J_p(\lambda, V) = \frac{e}{W}\left\{\mu_p F \int_0^W \delta_p dx - D_p \int_0^W \frac{\partial \delta_p}{\partial x} dx\right\} \quad (4)$$

where, p in the subscript denotes holes.

The V_{oc}, in tandem cell is taken to be equal to the summation of the V_{oc} of individual junction.

$$V_{oc} = \sum V_{oc(i)} \quad (5)$$

where, i is the number of junctions in the tandem cell ($i = 1, 2, 3, \ldots n$) and n is the number of junctions in cell. Thus, the V_{oc}, is given by

$$V_{oc} = \frac{nkT}{q} \times \ln\left(\frac{J_{pki}}{J_0} + 1\right) \quad (6)$$

where, k is the Boltzmann constant, J_0 is the saturation current density, n is the ideality factor, q is the electron charge density, and T is the temperature.

The transmitted light via top sub cell are calculated using

$$S(\lambda) = S_0(\lambda) \cdot \exp(-\alpha_{window}(\lambda) \cdot d_{window}) \cdot \exp(-\alpha_{absorber}(\lambda) \cdot d_{absorber}) \quad (7)$$

where, S_0 represent spectrum of incident light, α is the absorption coefficient and thickness of the cell material is d.

III. RESULTS AND DISCUSSION

Three different structures for tandem organic solar cells are investigated with the objective to improve the PCE (η). In all the three proposed structures, the impact of top and bottom sub-cell thickness and temperature on the V_{oc}, J_{sc}, FF and η is studied. To better understand the complete tandem structure, we have first investigated single junction cells as revealed in Fig. 2. The first structure shown in Fig. 2a is based on P3HT: PCBM material, which is the most common absorber material used in organic solar cell due to large bandgap of the donor polymer feature that improves the stability and match perfectly with solar spectrum [34]. The second cell shown in Fig. 2b is based on PTB7: PCBM, which is a most promising narrow bandgap polymer for harvesting light and has large absorption coefficient that increases the performance of the cell [35]–[37]. The hole transport layer (HTL) for both the cells is chosen as PEDOT: PSS due to good electrical conductivity [38]. The thickness of different layers of the cell are chosen as: P3HT: PCBM and PTB7: PCBM = 200nm, PEDOT: PSS = 40nm, while top and bottom electrode is chosen as 125 nm and 100 nm, respectively. Both the cells

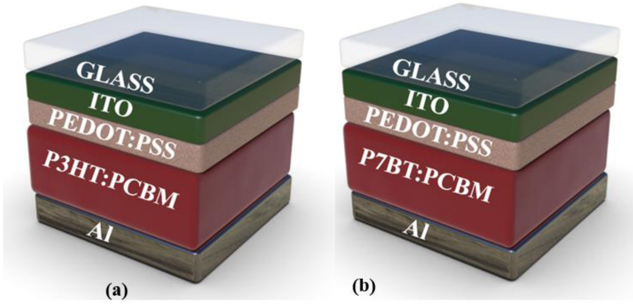

FIGURE 2. Single junction structure based on (a) P3HT:PCBM (b) PTB7:PCBM.

TABLE 1. Extracted photovoltaic parameters of single junction OSC.

Organic Material	V_{oc}(V)	J_{sc} (mA/cm^2)	FF %	η %
P3HT: PCBM	0.60	22.51	67.39	4.59
PTB7: PCBM	0.60	22.57	67.48	4.81

are simulated and the results are displayed in Table 1. The electrical parameters obtained for both the solar cells are appear to be approximately the same. However, such low efficiency is not suitable for most of the practical applications.

In order to counter the problem of low efficiency, we converted the single junction solar cell into multijunction, which is simply design of stacked materials that are optimized to create electric currents in response to different wavelengths of light. The double absorber layers will trap maximum photons compared to single absorber and thus, increase the absorption and efficiency of the whole cell. We first configured homo tandem structure by utilizing same absorber material (P3HT: PCBM) for top and bottom sub-cells as shown in Fig. 3. ITO and Al are used as top and bottom electrodes, while MoO3 is used as HTL layer. Furthermore, ZnO in the structure is used as tunnel junction, which will offer optically low-loss connection and very small electrical resistance between the two sub-cells. The electrical parameters extracted using the homo tandem cell are V_{oc} = 0.75V, J_{sc} = 22.74(mA/cm^2), FF = 66.9% and η = 11.48%, respectively. These values show that the double junction cell has boost the efficiency from 4.52% to 11.48%, indicating that the tandem approach is the best solution to enhance the performance of the cell in greater amount.

It is well known that the electrical parameters strongly depends on the thickness of the absorber layer in the top and bottom sub-cells [39]. Therefore, one can further improve the efficiency and other parameters by varying the thickness values. Figure 4a shows the influence of different top sub-cell absorber layer thickness on J_{sc} and V_{oc}. The results indicate that as the thickness of the absorber layer increases from 230-360 nm, the J_{sc} rises from 18.70-22.74 mA/m^2, while V_{oc} is slightly affected. This is because as the thickness of the absorber layer increases, the absorption of the incident photons also increases due to large area and generate more number of electron-hole pairs. Figure 4b shows the improvements in FF and η as the thickness value is changed.

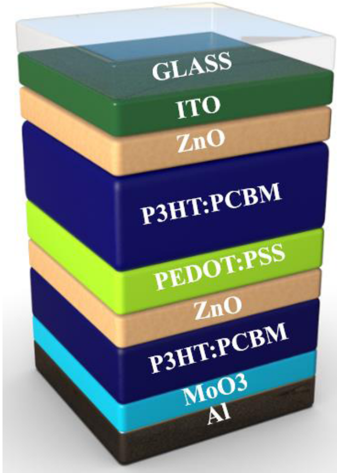

FIGURE 3. Schematic of homo tandem OSC structure based on P3HT:PCBM absorber material.

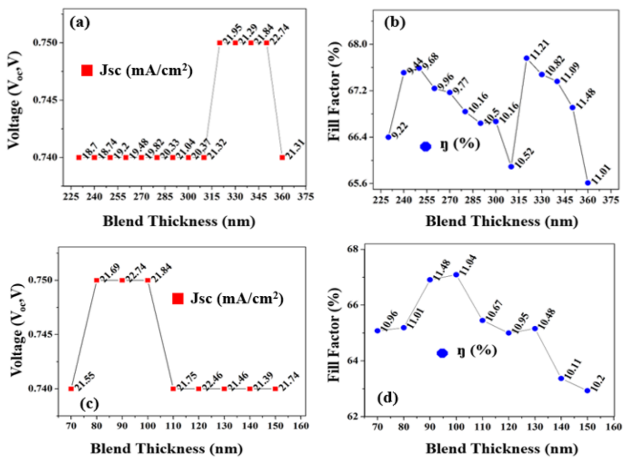

FIGURE 4. Influence of top sub-cell thickness on (a) V_{OC} and J_{SC}, (b) *FF* and η. Influence of bottom sub-cell thickness on (c) V_{OC} and J_{SC}, (d) *FF* and η.

The efficiency is enhanced for large thicknesses because it is directly linked with the absorption of incident photons, and one can generates and collects more carriers. However, for very large thickness values (360nm), the photo absorption remains the same, while collection efficiency decreases, resulting in low conversion efficiency η.

In Fig. 4, the effect of the bottom sub-cell thickness is also tested as this layer has the advantage to absorb low frequency photons, which usually penetrated from the top sub-cell. When the thickness is varied from 70-150 nm, the J_{sc} value first increases and then drops as shown in Fig. 4c, which in turn results in decrease η from 11.48% to 10.20% as shown in Fig. 4d. The efficiency drops with the absorber layer thickness is connected with increase in the series resistance of a solar cell. Therefore, the 90 nm thickness is found to be an optimum thickness for bottom sub cells to attain a good η.

The temperature influence on homo tandem cell is also studied as shown in Fig. 5. It is well known that high temperature has a negative influence on solar harnessing devices

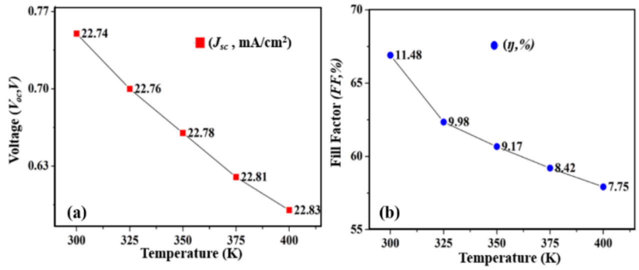

FIGURE 5. Influence of temperature on (a) V_{OC} and J_{SC}, and (b) on *FF* and η.

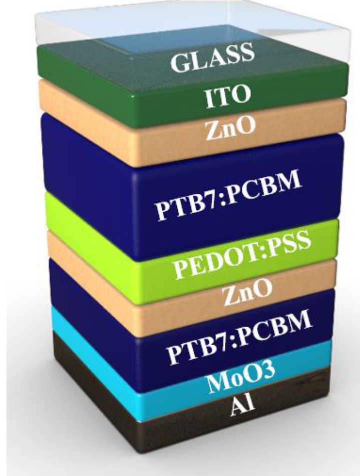

FIGURE 6. Schematic of homo tandem OSC structure based on PTB7: PCBM.

because bandgap E_g is not stable under high temperatures, which lead to higher recombination losses [40]. The same influence of excessive temperature is perceived on our proposed structure and the observation suggest that high temperature greatly decline the performance including V_{oc} and η. In Fig. 5a, J_{sc} slightly increases with temperature because the bandgap energy reduces, which results in greater electron–hole pairs generation. However, this increase in J_{sc} may give rise to thermalization losses, which significantly reduces the performance of the cell. The V_{oc} on the other hand highly drops from 0.76 V to 0.59 V due to its inverse relationship with temperature as given in equation (8).

$$\frac{d\left(V_{oc}\right)}{dT}=\frac{1}{T}\left(V_{oc}-\frac{E_g}{q}\right) \tag{8}$$

A. HOMO TANDEM PTB7:PCBM

We next designed homo tandem cell from low bandgap organic PTB7: PCBM material as shown in Fig. 6. The extracted electrical parameters for this cell are: $V_{oc}=0.75$ V, $J_{sc}=25.40$ mA/cm^2, $FF=66.94\%$ and $\eta=12.90\%$, respectively. In this case, η is improved from 4.79% (single junction) to 12.90%.

The effect of absorber layer thickness for the top and bottom sub-cells are also examined in this case. Figure 7 shows

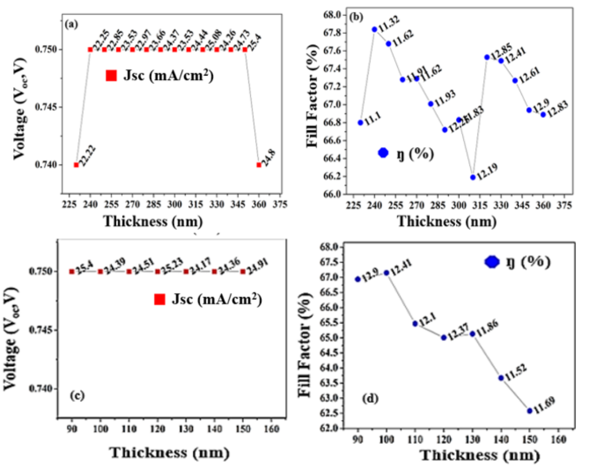

FIGURE 7. **Influence of top sub-cell thickness on (a) V_{OC} and J_{SC}, (b) FF and η. Influence of bottom sub-cell thickness on (c) V_{OC} and J_{SC}, (d) FF and η.**

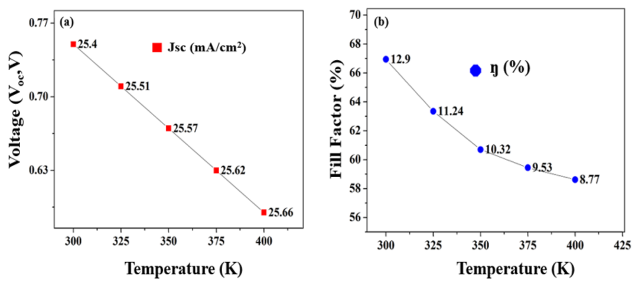

FIGURE 8. **Influence of temperature on (a) V_{OC} and J_{SC}, and (b) on FF and η.**

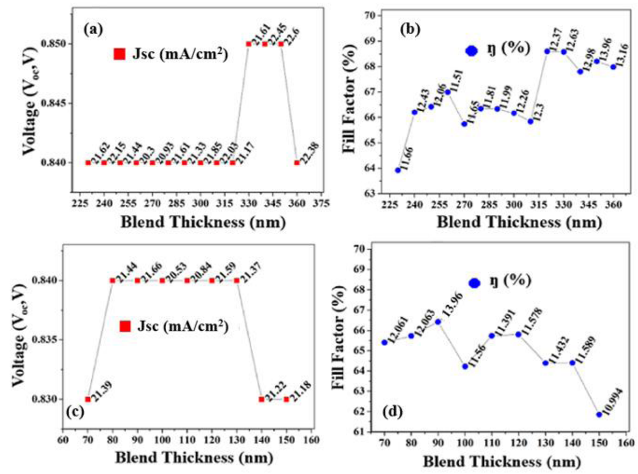

FIGURE 9. **Influence of top sub-cell thickness on (a) V_{OC} and J_{SC}, (b) FF and η. Influence of bottom sub-cell thickness on (c) V_{OC} and J_{SC}, (d) FF and η.**

the variation in photoactive absorber layer thickness on J_{sc}, V_{oc}, FF and η, respectively. As the thickness of the photoactive absorber layer increases, the J_{sc} and V_{oc} also increases in the same manner as in the previous case. However, here, J_{sc} values are found slightly greater compared to previous case due to high absorption coefficient i.e., 67237cm^{-1}. Similarly, in Fig. 7b, the η rises from 11.10% to 12.90% as the thickness increases. The η recorded for this structure is also more compared with the previous case. Figure 7c and 7d, which indicates the thickness variation of bottom sub-cell is also similar to the previous case. However, the J_{sc} and η values are found higher in this case. Therefore, the above calculations are generally similar for all tandem solar cells [41].

The influence of temperature in this case is also studied as shown in Fig. 8. As discussed earlier, excessive high temperature has pessimistic impact on the electrical parameters, which leads to poor performance of the cell.

B. HYBRID TANDEM SOLAR CELL

To further improve the performance of the tandem solar cell, hybrid absorber layers with different bandgaps are mostly used as it is possible to extract efficiency beyond the Shockley-Queisser limit [42]. We designed a hybrid tandem cell as shown in Fig. 1, where the top sub-cell absorber layer is made of P3HT:PCBM, accountable for the absorption of high energetic photons, while the bottom absorber layer is made of PTB7:PCBM, responsible for the absorption of low energy photons. The choice of introducing PTB7:PCBM for bottom sub-cell layer is like a corner stone that plays a pivotal role, as the absorption coefficient of PTB7:PCBM is $\alpha = 67237$ cm^{-1}, which is greater than P3HT:PCBM whose absorption coefficient is $\alpha = 52925$ cm^{-1} [36]. Therefore, by using different absorption coefficients with different bandgap values essentially help to absorb incident photons in large quantities, resulting in a high conversion efficiency η. The extracted electrical parameters for the hybrid cell are: $V_{oc} = 0.85$ V, $J_{sc} = 22.60$ mA/cm^2, $FF = 68.21\%$ and $\eta = 13.96\%$, respectively. These values are higher than the homo tandem cells, indicating that the hybrid approach is more useful for practical applications.

To obtain the optimum values for the electrical parameters, we varied the thickness of both the absorber layers. For the top sub-cell, the thickness of P3HT: PCBM is altered from 230-360 nm, while the bottom sub-cell absorber layer (PTB7: PCBM) thickness is fixed at 90nm. Figure. 9a indicates that when the absorber thickness of the top sub-cell (P3HT: PCBM) is increased from 230-350nm, the J_{sc} value essentially increases and attain the high value of 22.6mA/cm^2 at 350 nm, which is higher than the previously investigated homo structures. In Fig. 9(b), the highest η of 13.96% is achieved at 350nm thickness, which to our knowledge is the highest efficiency for organic solar cells. Figure 9(c) shows the effect of bottom sub-cell thickness, which is varied from 70-150 nm and the results indicate that the optimum thickness for bottom sub-cell is 90 nm, which supports highest η as shown in Fig. 9(d). Therefore, the optimal thickness for top and bottom sub-cell for achieving high η of 13.96% is 350nm and 90nm, respectively.

Next, we replaced PEDOT:PSS in the top sub-cell with MoO_3(molybdenum trioxide), ZnO (zinc oxide), and TiO_2 in the bottom sub-cell with C_{60} (buckminsterfullerene), MoO_3, Ca (calcium) of the proposed hybrid tandem cell. The results

TABLE 2. Extracted performance parameters obtained by replacing PEDOT: PSS and TiO_2 in hybrid tandem cell.

Parameters	PEDOT:PSS (Top Sub-Cell)			TiO_2 (Bottom Sub-Cell)			
	PEDOT:PSS	MoO_3	ZnO	TiO_2	C_{60}	MoO_3	Ca
V_{oc} (V)	0.85	0.69	0.69	0.85	0.68	0.68	0.68
J_{sc} (mA/m^2)	22.60	24.10	25.27	22.60	22.88	23.09	22.83
FF %	68.21	60.33	59.99	68.21	60.42	60.42	60.64
η %	13.96	10.05	10.51	13.96	9.62	9.62	9.56

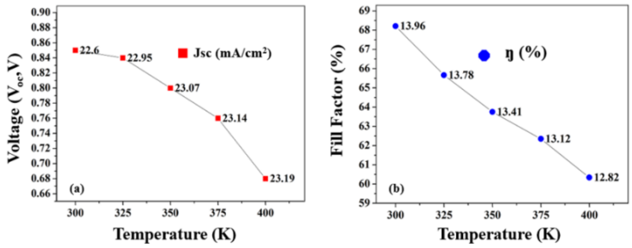

FIGURE 10. Influence of thickness on (a) V_{OC} and J_{SC}, and (b) *FF* and η.

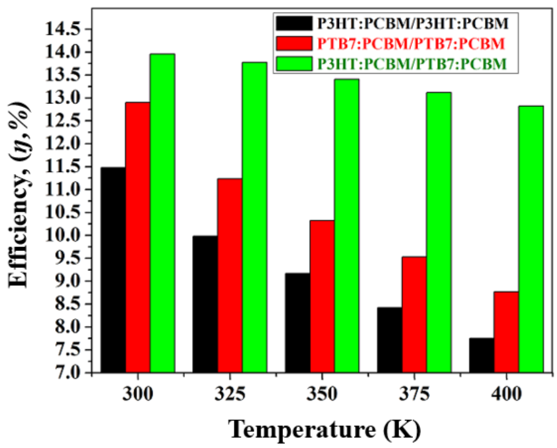

FIGURE 11. Temperature effect on the efficiency of homo and hybrid tandem solar cells.

are summarized in Table 2, which indicates that the materials we selected before are best for tandem cell because of high efficiency.

The high temperature effects on the performance of hybrid cell is also tested as shown in Fig. 10. It appears that the performance of the cell is reduced by increasing the temperature like other kind of solar cells. In Fig. 11, we have compared the efficiency of all the proposed tandem cells versus temperature. It is found that for high temperature (400K), the efficiency of hybrid cell is far better than the homo tandem cells. This indicates that hybrid tandem cell is more appropriate for practical applications.

Eventually, the efficiency of the hybrid solar cell is compared with the existing solar cells in the literature as revealed in Table 3. The results show that our proposed structure holds the highest conversion efficiency compared to others.

TABLE 3. Comparison of different solar cells with current study.

Reference	Structure	η (%)
[43]	P3HT:ICBA/PBDTT-DPP	8.62
[44]	a-Si:H p-i-n/PDTP-DFBT:$PC_{71}BM$	10.50
[45]	P3TEA:SF-PDI_2/ P3TEA:SF-PDI_2	10.8
[46]	P3TEA:FTTBPDI4/PTB7-Th:IEICS-4F	10.5
Current Study	P3HT:PCBM/PTB7:PCBM	13.96

IV. CONCLUSION

We theoretically analyzed three different kinds of organic tandem cells and compared their performance parameters. The homo tandem cell based on P3HT: PCBM material supports slightly low efficiency compared to PTB7: PCBM based cell because of small absorption coefficient. However, the optimized hybrid tandem solar cell, which is a combination of both absorber materials used in homo cells exhibit high efficiency of 13.96% because the top active layer absorb high frequency photons and bottom layer absorb low frequency photons. Moreover, the hybrid cell shows more stability towards high temperatures and offers highest conversion efficiency by comparing with the existing organic solar cells.

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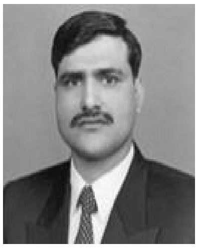

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