An Efficient Non-Toxic and Non-Corrosive Perovskite Solar Cell

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ABSTRACT This article reports a computational comparison modelling of two types of thin film perovskite solar cells which includes a standard lead based perovskite solar cell and tin based solar cell. The lead based perovskite solar cell shows better performance, however, they are not sustainable for a longer period of time due toxic materials, which are not environmental friendly and corrosive materials that react with perovskite layer and slowly corrode it, reducing the efficiency and badly affecting the physical state of the cell. Therefore, we proposed a tin based perovskite solar cell, which contains non-toxic and non-corrosive materials. Results indicate that the standard lead based perovskite solar cell show only 2.63% more efficiency as compared to non-toxic one. In addition, the stability under different operating temperatures for both kinds of cells have been tested and it is observed that the lead based structure is 3.36% more stable than tin based structure. These analysis show that the tin based cell performance is not much lower than the lead based cell. Therefore, by considering the environmental and stability issues, the preferable choice is the tin based perovskite solar cell.

INDEX TERMS Tin (Sn), Lead (Pb), Perovskite, optimization, and stability.

I. INTRODUCTION

In the latter half of the decade, Perovskite solar cells have caught the attention of the research community, due to its enormous advantages over other semiconducting materials that can be utilized for Thin Film technology (TFT). Moreover, the ability to harvest high power conversion efficiency (PCE) makes this material an epoch candidate for solar cells. Many kinds of perovskite materials are available in the market, but they mostly contain some toxic materials like Lead halide and Bromide halide[1]. These toxic materials are harmful to the environment and produce environmentally unfriendly effects during manufacturing. These toxic materials are not environment friendly and their manufacturing could be a potential environmental hazard. In this context, non-toxic materials are available that can be molded with Methylammonium. Tin (Sn) is one such non-toxic material that can be used instead of Lead (Pb) in perovskite solar cell, which has attracted the PV research market because of its outstanding electrical and optical properties [2], [3].

Additionally, Perovskite solar cells have high absorption coefficient, low manufacturing cost, low recombination kinetics and high mobility [4]. The formula of Perovskite solar cells for the compound ratio is ABX$_3$, where A, B represents Methylammonium (CH$_3$NH$_3$), is divalent cations and X represents cations. This ABX$_3$ (Perovskite) layer is sandwiched between the Electron transport layer (ETL) and Hole transport layer (HTL), covered by the top and bottom electrodes [5]. For the top electrode, transparent conductive oxide (TCO) is used whereas, for the bottom electrode, usually metal is utilized [6], [7]. The working principle of PSC is displayed in Fig. 1.

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Different approaches have been numerically investigated to improve the cell photovoltaic performance parameters. J. Duan et al., investigated inorganic perovskite solar cell and obtained an efficiency of 10.14% [8]. N. Kumari et al., obtained an efficiency of 7.39% from perovskite solar cell by using 4-tertbutylpyridine as stabilizer [9]. L. Zhang et al., implemented general strategy of solution engineering and reported an efficiency of 12.5% [10]. R. Singh et al., reported an efficiency of 18.7% from perovskite solar cell by utilizing PCBM as an additive [11]. H. Sun et al., recorded an efficiency of 20.1% from perovskite solar cell by using novel transparent conductive mesoporous layer [12]. H. Baig et al., obtained an efficiency of 21.6% from PSC by utilizing mesoporous ETL composed of TiO\textsubscript{2}/SnO\textsubscript{2} [13]. Y. Dou et al., reported single junction ST-PSC and recorded an efficiency of 14.69% [14]. S. Das et al., fabricated perovskite solar cell and obtained an average efficiency of 16.34% [15]. C. Wang et al., reported an efficiency of 11.4% from lead free PSC [16]. S. Abdelaziz et al., numerically modelled and calculated an efficiency of 14.03% from formamidinium tin based perovskite solar cell [17]. Q. Fu et al., fabricated stable tin based perovskite solar cell and obtained an efficiency of 7.3% [18].

Herein, we reports numerical modelling of thin film perovskite solar cells based on Toxic and Non-Toxic material. The comparative analysis between both the structures ((i) Glass/FTO/TiO\textsubscript{2}/Tin based perovskite layer/Cu\textsubscript{2}O/Cu, and (ii) Glass/FTO/TiO\textsubscript{2}/Lead based perovskite layer/Cu\textsubscript{2}O/Cu) have been performed to highlight the role of lead free and non-corrosiveness structure for thin film technology. Results show that the corrosive and toxic free perovskite structure exhibit slightly less performance than the standard lead based perovskite solar cell. This indicates that the proposed tin based solar cell can be commercialized due to eco-sustainability.

II. DEVICE MODELLING AND FRAME WORK
The proposed structures illustrated in Fig. 2, are composed of different layers. The illustrated architecture started from the protection layer of Glass having thickness of 60 nm. For the top electrode, FTO is used, having thickness of 125 nm. The FTO provides high transparency for the under lying layers and shows high thermal stability [19]. For ETL, titanium dioxide (TiO\textsubscript{2}) is considered having thickness of 20 nm. TiO\textsubscript{2} layer is considered as one of the most effective layer for ETL block, as it has high mobility, high transparency, and have excellent thermal stability. Moreover, it can also quench more electrons for the bottom layers [20]. Next to the ETL stack, photoactive layer is introduced (Sn and Pb based Perovskite) having variable thickness. The Sn based material is
non-toxic [21] and can be probable more preferable over toxic Pb based perovskite because the toxic materials produce harmful effect on the environment during manufacturing [22]. Next for the flow of holes, HTL layer composed of Copper Oxide (Cu$_2$O) having thickness of 540 nm is stacked as its can be utilized to reduce the amount of Cu, which has been used for the bottom electrode. Through this simulation, the optimization for window, active and hole transport layer are optimized for efficiency enhancement. If the thickness of the cell is not optimized, it give rise to certain problems such as series resistance, defect in state densities, recombination loses and decline in diffusion duration [23], [24]. Moreover, electrons cannot travel long distance because of the short life span [25], [26]. Thus, to counter these problems there is a dire need to optimize the cell with improved PV parameters.

For the simulations, GPVDM [27] software is utilized, which is a free online platform for photovoltaic research community featured with both the models (optical + electrical). The software is based on the different standard approaches including the drift diffusion model, electrical mesh points, equation of continuity and poisson equation. Furthermore, for the recombination and trap states, Shockley Read Hall (SRH) formalism is used which helps in understanding the rates at which the electrons and holes can be escaped and captured in carrier traps. To understand the core of the device, the functionality can be understood by the orbitals energies such as LUMO and HUMO. The profile across the device for

- **TABLE 1.** Input parameters.

| Parameters          | ETL (TiO$_2$) | Pb based Perovskite | Sn based Perovskite | HTL (Cu$_2$O) | Cu   |
|---------------------|---------------|---------------------|---------------------|----------------|------|
| Thickness (nm)      | 20            | 450                 | 450                 | 540            | 180  |
| Band Gap Energy $E_g$ | 3.3          | 1.5                 | 1.3                 | 2.0            | 2.5  |
| Hole Mobility $\mu_h$ | $10 \times 10^{-4}$ | $50.0 \times 10^{-4}$ | $1.6 \times 10^{-4}$ | $4.55 \times 10^{-4}$ | $5 \times 10^{-2}$ |
| Hole Density Of State | 0          | $9.1 \times 10^{26}$ | $1 \times 10^{24}$ | $1 \times 10^{25}$ | $3 \times 10^{28}$ |
| Electron Mobility $\mu_e$ | $20 \times 10^{-4}$ | $50.0 \times 10^{-4}$ | $1.6 \times 10^{-4}$ | $176 \times 10^{-4}$ | $43.4 \times 10^{-4}$ |
| Electron Affinity   | 4.0           | 3.93                | 4.17                | 2.9            | 7.72 |
| Electrons Density of State | $1 \times 10^{22}$ | $1.3 \times 10^{26}$ | $1 \times 10^{24}$ | 0              | $3 \times 10^{28}$ |
| Relative Permittivity | 9.0         | 3.0                 | 8.2                 | 5.0            | 6.0  |

**FIGURE 3.** Optimization of ET Layer Thickness against the functionality of device for (a) $J_{sc}$ (b) $V_{oc}$ (c) $FF$ and (d) $PCE$. 
FIGURE 4. Optimization of Active Layer Thickness against the functionality of device for (a) J_{sc} (b) V_{oc} (c) FF and (d) PCE.

the potential of space charge dependent is based on the well-known poisson equation which can be expressed by Eq. 1.

$$\frac{d}{dx} \varepsilon_o \varepsilon_r \frac{d\varphi}{dx} = q (n-p)$$ (1)

For the device charge conversion, bipolar drift diffusion model is utilized, and can be written as Eq. 2 and Eq. 3.

$$\frac{\partial J_n}{\partial x} = q \left( R_n - G + \frac{\partial n}{\partial t} \right)$$ (2)

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

The Fill factor (FF) of the device can be calculated by Eq. 4.

$$FF = \frac{J_{mp}V_{mp}}{J_{sc}V_{oc}}$$ (4)

The device PCE can be obtained by the Eq. 5

$$PCE (\%) = \frac{V_{oc} \cdot J_{sc} \cdot FF}{P_{input}} \times 100$$ (5)

All the simulations have been carried out under the standard testing conditions, where the temperature of the device is set to constant at 300K throughout the modelling and optimization. The input electrical parameters are displayed in Table 1.

III. RESULTS AND DISCUSSION
Optimization of the effective layers in terms of materials and thickness puts an outmost impact on the device performance [28]–[30].

A. OPTIMIZATION OF TITANIUM DIOXIDE (TiO_2) AS ELECTRON TRANSPORT LAYER FOR DEVICE IMPROVEMENT
Electron Transport Layer (ETL), in solar cells is one of the most important and crucial layer, as it is the first step for light to enter in the cell material. The selection of the ETL is an important step while designing the cell architecture. Such devices result in low performance if the selected material is not compatible with the proposed architecture. After selecting the appropriate material for the ETL stacked, the optimization is the next door step for the designing of the cell. If the material is utilized without optimization, it may result in low efficiency and increase the manufacturing cost of the device. In this proffer case, the thickness of the ETL is modulated and optimized at room temperature. The thickness is varied from 10-100 nm while keeping the active layer (Sn) constant at 400 nm. As the thickness increases from 10-20 nm, the J_{sc} value rises from 22.31-22.37 mA/cm², V_{oc} increases from 0.887-0.891(V), FF increases from 83.61-83.69% and PCE increases form 16.47-16.54%. However, further increase in
the thickness of the window layer resulted in the decline of all the performance parameters as shown in fig. 3 (a-d), respectively. These recorded parameters further suggest that 20 nm has been found to be adequate with the proposed structure (Sn) to achieve the highest efficiency of 16.54%.

Next, Sn is replaced with the toxic lead based (Pb) perovskite material and the same strategy is implemented in this structure as well. Here again, a thickness of 20 nm for TiO$_2$ layer is found to be the optimum value as shown in Fig. 3 (a-d), respectively. In this case, the highest PCE of 17.89% is obtained along with other photovoltaic parameters such as $J_{sc} = 22.71$ mA/cm$^2$, $V_{oc} = 0.84$V, and $FF = 87.89\%$. As an upshot, if one has to obtain high $V_{oc}$, than the Sn based structure can be utilized.

**B. OPTIMIZATION OF TIN AND LEAD BASED PEROVSKITE MATERIAL AS LIGHT HARVESTING LAYER FOR DEVICE IMPROVEMENT**

After optimizing the window layer, the next crucial step involved in the design of the cell is the optimization of the photoactive layer. This light harvesting layer is one of the most important and critical parameter as all the support of the device performance lying on this layer [31]. Optimization of this critical layer is another milestone to achieve, as it boost the device performance by absorbing large amount of light, which further enhance the pair of excitons in the active region [32]. Two different kind of active materials have been tested and optimized with the objective to put forward such an optimized design of the cell, which can be utilized for future applications with minimum harmful effect on the environment during manufacturing. The two proffers materials are Sn based perovskite and Pb based perovskite. Sn is non-toxic in nature, whereas, Pb is toxic [33], [34]. Both the materials have been equally monitored and numerically reckon to provide maximum high electrical photovoltaic parameters. The thickness of both the materials altered between 100-1000 nm. The highest efficiency for both the materials obtained at 450 nm and shows the identical behavior. More obviously, Sn based device trail than Pb based device. For the Sn based layer, the recorded high values of the photovoltaic parameters are: $J_{sc} = 22.82$ mA/cm$^2$, $V_{oc} = 0.87$V, $FF = 82.49\%$ and $PCE = 21.22\%$, while for the Pb the obtained parameters are: $J_{sc} = 24.18$ mA/cm$^2$, $V_{oc} = 0.84$V, $FF = 87.88\%$ and $PCE = 23.82\%$, respectively. Thus, Pb is found to be 2.6% more efficient than Sn. However, the $V_{oc}$ in Sn based device is much better than the Pb counterpart. In addition, the Pb based devices, show high mobility, high charge carriers, and put high inherent for hole/electron states of density as compared to Sn, which resulted in the high conversion efficiency. The FF of the Sn-based device shows a linear decrease, while this decrease is less in the Pb-based device. The FF of the device is basically shows how effectively the flow of electron/hole is
C. OPTIMIZATION OF COPPER OXIDE (Cu$_2$O) AS HOLE TRANSPORT LAYER FOR DEVICE IMPROVEMENT

After optimization of the active layer, HTL is introduced for the purpose of improving the flow of holes in the cell and blocking electrons to enter the hole majority area. Different materials have been utilized in the literature with perovskite solar cells, such as Spiro-OmeTAD [36] and P3HT [37]. Here, we have used copper oxide (Cu$_2$O) as HTL [38] and optimized it in both the structures. Surprisingly, the performance of the cell boosted by giving the highest PCE of 21.45%, $J_{sc}$ of 23.14mA/cm$^2$, $V_{oc}$ of 0.885(V), and $FF$ of 82.59% with Sn based structure, while with the Pb based structure the PCE of 24.08%, $J_{sc}$ of 24.45mA/cm$^2$, $V_{oc}$ of 0.854(V) and $FF$ of 88.02%, is attained. The optimized thickness of 540 nm is adequate to achieve high performance parameters for both the structures as shown in Fig. 5 (a-d), respectively. This further suggest that, Cu$_2$O can be considered as a replacing layer to the typical HTL such as P3HT and Spiro-OmeTAD. The amount of Cu in Cu$_2$O can be utilized for the bottom electrode going without losses. Moreover, the bell shaped in the graphs for power conversion efficiency is evident that as the thickness increases, the exciton pairs increases [35], which may also results in recombination and curved occurred. Further increase in the thickness for both the materials resulted in the decline trends.
and may be considered as a strong candidate to reduce the amount of cost for the cell. In addition, Cu is non-corrosive material and improve the stability and life of the cell [39]. The other typical materials for the bottom electrode are silver (Ag), and aluminum (Al), which produce corrosiveness that might harm the active material that ultimately decrease the performance of the cell. In this scenario, Cu2O can be considered a strong candidate for the proffer structures as it provides an enhanced stability for the HTL layer as well as for the bottom electrode [40]. Thus, the recorded parameters with and without Cu2O are summarized in Table 2 and structure with Cu2O show high PCE as compared to without HTL.

D. TEMPERATURE ANALYSIS

Solar cells are extremely sensitive to high temperature. The device performance decreases with the increases in the temperature. As shown in Fig. 6, all the electrical parameters exhibit a downward trend and low efficiency as the temperature increases. Jsc is the only PV parameter, which slightly increases with the increase in the temperature. This increase is extremely small and does not contribute much in the enchantment of cell performance. However, this small gain in the value of Jsc occur due to instability of the energy band gap of the active layer. The bandgap of the materials shows instability under high temperature [41] due to which its width decreases and pairs of e/h are created in large amount, which makes this small gain in the section of Jsc. In this case, when the temperature of the device varied from 300-400K, the PCE decreases from 21.45-20.16% for Sn based structure and 24.08-23.52% for Pb based structure. Moreover, the stability of the cell decreases by 1.29% for Sn based and 0.56% for Pb based. Thus, suggesting that Pb based perovskite solar cell is more stable under such conditions. Moreover, Voc and FF also decreases because of reverse saturation current. The Voc in terms of temperature is expressed as [42].

\[
\frac{d(V_{oc})}{dT} = \frac{1}{T} \left( V_{oc} - \frac{E_g}{q} \right)
\]

IV. CONCLUSION

In conclusion, we computationally optimized perovskite solar cell based on toxic and non-toxic materials, featured with optimized ETL and HTL layer. For the ETL, TiO2 is found to be optimized at 20 nm and for HTL, Cu2O is observed to be optimized at 540 nm. The structure with HTL appears to be more efficient as compared to structure without HTL. Thus, an enhancement gain of 0.23% and 0.26% is observed with Sn and Pb based structure. Moreover, optimization of toxic and non-toxic perovskite layer is performed with the proposed structure and both the proffer structures shows a similar trend after attaining the optimum thickness of 450 nm. Thus, toxic lead based perovskite material is found to be more efficient by delivering the highest η of 24.08%, which is 2.63% more than the non-toxic perovskite layer that delivers the highest η of 21.45%. Additionally, to check the stability and sensitivity, the cells have been tested on different temperatures and the obtained results reveal that the lead based perovskite is 3.36% more stable as compared to tin based perovskite. Although, the non-toxic cell is slightly low in performance when compared to the toxic one, however, it has the potential and ability to replace the existing lead based perovskite solar cells.

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